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Fertilizers and Pesticides Impact on Surface-Active Substances Accumulation in the Dark Gray Podzolic Soils

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ABSTRACT

The article reports on a study that examined the impact of agrochemicals on the levels of surfactants in soil. Specifically, the study found that the use of mineral fertilizers and pesticides led to an increase in the levels of anionic surfactants (ASA) in the soil. Furthermore, the simultaneous application of fertilizers and pesticides had a greater effect on ASA levels than either factor alone. The use of pesticides also led to an increase in non-ionic surfactants (NSA), while the use of fertilizers resulted in a decrease in NSA levels. The study also found that the increase in the levels of mobile forms of key nutrients in the soil was associated with the accumulation of ASA in lower layers of the soil profile.The amount of alkaline hydrolyzed nitrogen under the low protection system increased by 3.0–23.2 mg kg^{-1} soil, mobile phosphorus by 14.0–144.0 mg P_2O_5, and exchangeable potassium by 9.0–222.0 mg K_2O per kg soil, compared to the control. With the complex use of fertilizers and pesticides in one block, a trend of increasing mobile forms of nutrients in the soil was observed. The distribution of ASA amount in the soil profile is descending. The clear presence of ASA was established only in the soil layer of 0–40 cm. An increase of ASA content in the soil due to the use of agrochemicals and fertilizers is observed up to a depth of 60–80 cm. Using biological elements in agriculture significantly reduces the amount of these substances in the soil profile.

Keywords: mineral fertilizers; soil; pesticides; anionic surfactants; non-ionic surfactants; crop rotation; biogenic elements; agroecosystem.

INTRODUCTION

The need to increase the gross collection of plant products requires changes in the technologies for growing agricultural crops (Havryliuk et al. 2022a, 2022b). This led to the transition to intensive technologies, which involved the widespread use of fertilizers and plant protection agents (chemical substances for combating pests, plant diseases, and weeds). Systematic use of pesticides turned them into a stable ecological factor that changes and shapes macro- and microcenoses (Hrabovska et al., 2011). After all, the entry of persistent pesticides into the air flows leads to the global migration of toxic drugs. A comprehensive survey of various natural objects of the agro landscape, including grown agricultural products, showed that the use of pesticides does not pollute individual components, but all interconnected natural environments and objects (Farias et al., 2021; Rasheed et al., 2020). Pesticide residue monitoring data in the USA confirm their presence in almost all controlled objects of agrophytocenoses. One of the sources of rivers and reservoirs pollution is pesticides removal from agricultural lands by surface runoff (Siyal et al., 2020; Jimoh & Johnson, 2019; Palmer & Hatley, 2018). The higher is pesticides solubility in water, the more mobile the toxicant in the soil and the greater danger of its vertical downward migration (Bansal, 2011; Saleh et al., 2022).

The introduction of a large number of pesticides, growth regulators, into the agroecosystem can lead to the accumulation of residues of these compounds or their metabolites in the biosphere (Badmus et al., 2021; Silva, et al., 2019; Zhao et al., 2015; Pérez & Eugenio, 2018). In some cases, it is metabolites that have greater resistance to decay, toxicity, mutagenicity, and carcinogenicity compared to the original xenobiotics. These facts explain the need to study the possibility of soil self-cleaning from agrochemical residues (Orton et al., 2013; Masia et al., 2015; Pose-Juan et al., 2015; Qu et al., 2016; Chiaia-Hernandez et al., 2017; Hvezdova et al., 2018).

According to the research of a number of scientists, it is soil microorganisms that play the role of active pesticides destroyers, subjecting them to biological transformation, while indicators of the number of microbiological communities and their activity can act as criteria for soil monitoring (Kruglov et al., 1980; Zvyagintsev et al., 1992, Symochko, 2020). Researchers have described a number of cases of negative herbicides' effects on the number of bacteria (Kuyan, 1977; Deshmuk & Shrikhande, 1977; Sherry, 1994). Research by Ananyeva (1993) shows that the rate of pesticide decay in the soil increases with their regular use because as a result of the adaptation process of the soil microbial community microorganisms are capable of accelerating the biodegradation of pesticides accumulate. According to experts, as a result of the systematic use of pesticides, not only the biological, but also the agrochemical and physicochemical properties of the soil change, and the effect of the pesticide on soil fertility depends on the applied dose of the drug, the temperature, and humidity of the environment. Thus, in the studies of Bliev & Melnikova (1986), the herbicide glyphosate application at a dose of 20 kg ha^{-1} in field conditions led to an increase in the content of total nitrogen and humus in the soil due to the nonhydrolyzed residue. At the same time, glyphosate had no significant effect on the content of mobile forms of phosphorus and potassium in the soil. Long-term application of soil herbicides causes activation of organic matter decomposition, which reduces the content of humus and total nitrogen in the soil and increases the content of NO_3 (Butenko et al., 2020; Tanchyk et al. 2021).

It has been proven that pesticides, which are widely used in intensive agriculture, are a source of an additional influx of surface-active substances (surfactants) into the agroecosystem, as a mandatory component of pesticides, and represent a group of products of organic synthesis. Peculiarities of the molecular structure of surfactants determine their two main properties: adsorption from the solution on the surface of the interface and the formation of large aggregates (micelles) in the solution (Prodanchuk & Mudriy, 2000)

According to research data from the Institute of Ecohygiene and Toxicology named after L. I. Medved, the share of SA in pesticides, mineral fertilizers, and other agrochemicals reach 10%. Anionic and non-ionic surfactants are used to improve the physical properties of mineral fertilizers. By introducing surfactants into the composition of chemical preparations, an increase in the solubility and selectivity of pesticides is achieved. It has been proven that a load of surfactants on the environment due to the use of pesticides, mineral fertilizers, other agrochemicals, and irrigation of land with wastewater amounts to 20–25% of the total input.

It is obvious that the use of surfactants will continue to expand in agriculture. As a result of their application, the seed germination and yield of agricultural crops increases, the labour intensity of plant care decreases, and the effectiveness of pesticides and mineral fertilizers increases (Litvinova et al., 2020, 2021). In recent years, technologies involving the use of natural surfactants as plant growth stimulants, bactericidal and fungicidal drugs have become widespread (Prodanchuk & Mudriy, 2000). Surfactants reduce soil density, as a result of which its moisture-holding capacity, aeration improves, and the possibility of a surface crust formation decreases. Adsorbing on the walls of soil capillaries, surfactants contribute to the formation of menisci of reverse curvature, thereby reducing filtration losses of moisture in the soil. Soils have a high adsorption capacity for surfactants. Studies by Borneff (1974) indicate that non-cultivated soil contains surfactants in the amount of 1–10 $mg \cdot kg^{-1}$, and in the territories that are under cultivation, surfactants reach 100 mg kg^{-1}. Surfactant sorption depends on the granulometric composition of the soil and increases from sandy to loamy soils. Sandy soils are capable of holding up to 25% of exogenous anionic surfactants, and loamy soils up to 90% (Eliseev & Kucher, 1991; Hanislamova et al., 1988). The filtration of surfactants to the greatest extent occurs in soils of light mechanical composition and significantly less in loams.

The works of Ukrainian researchers proved that surfactants affect biological soil activity: changes in the quantitative and qualitative composition of the soil microflora were noted (Voloshchenko & Mudryiy, 1991). The nitrification capacity in all variants with the addition of surfactants was higher than in the control, and increasing the surfactant load from 100 to 158 g cm^2 inhibited the cellulose-degrading activity of microorganisms. At the same time, there was a further accumulation of detergents in the soil, and their concentration became generally toxic to microorganisms.

Thus, the widespread use of pesticides in agricultural crops growing has turned them into a stable stressor in the agrobiotope. In addition to ensuring the necessary phytosanitary condition of crops, pesticides serve as a source of entry of ecotoxicants into the agroecosystem (Vasylenko et al., 2021; Havryliuk et al. 2022a, 2022b, Ivanova et al., 2022). At the same time, it is necessary to understand that it is important not only the pollution by pesticides, their decomposition products, and related substances (surfactants) but also the reaction of the biocenosis to these stress factors. Thus, to this day, the question of pesticides' influence on the state of soil microorganisms, and therefore on soil fertility indicators related to the activity of the microbial coenosis, remains insufficiently elucidated.

MATERIALS AND METHODS

Field research was carried out in a long-term stationary experiment, established in 1987 on a dark-grey podzolic soil. Crop rotation in the experiment was grain-tilled: peas, winter wheat, winter rape, spring barley, and corn for grain. The sowing area is 42 m^2, and the accounting area is 25 m^2. The repetition in the experiment is quadruple. When determining the long-term effects of chemicals and their residues on effective soil fertility, peas of the 'Gotivskyi' variety were used as a test crop.

The following were determined in the soil samples:

- humus – according to Tyurin (the method is based on the oxidation of the carbon of humus substances to CO_2 0.4 by the solution of potassium dichromate ($K_2Cr_2O_7$), prepared on sulfuric acid diluted in water 1:1. According to the amount of the chromium mixture that was used for the oxidation of organic carbon judging by its quantity) (DSTU 4289:2004, 2005)
- hydrolyzed nitrogen by alkaline – according to Kornfield (the method consists in hydrolyzing a weight of soil by 1H NaOH solution in a thermostat at 28 °C in a Conway cup) (DSTU 7863:2015, 2016).
- mobile phosphorus – by the Chirikov method (the method is based on the extraction of mobile compounds of phosphorus and potassium from one weight of soil by 0.5M solution of CH_3COOH at a ratio of soil:solution = 1:2.5 with the subsequent determination of phosphorus on a photoelectrocolorimeter) (DSTU 4115-2002, 2003).
- mobile potassium – by the Maslova method (the method is based on the extraction of potassium from the soil absorption complex by 1M solution of CH_3COONH_4 at a ratio of soil:solution = 1:10. (DSTU 7907:2015, 2016).

Definition of SA class. Pour 8 ml of methylene blue solution into a test tube (25 ml), add 5 ml of chloroform and drop by drop a 0.05% aqueous solution of a known anionic substance, vigorously shaking after adding each portion. The test tube is left to stand until the solution separates into two layers and it is observed how the blue colour of the upper aqueous layer, as the solution of a known anionic surfactant is added, gradually passes into the chloroform lower layer, which is due to the formation of a complex of methylene blue soluble in chloroform with a known anionic substance. The solution of the latter is added until the colour of the aqueous and chloroform layers equalizes and becomes blue. Then add 2 ml of an aqueous solution of the studied surfactant and shake vigorously. If, after settling, the aqueous layer becomes discoloured, and the chloroform layer becomes dark blue, then the substance under study belongs to anionic surfactants. If the reverse pattern is observed (the aqueous layer is intensely coloured, and the chloroform layer is decoloured), then the surfactant is cationic. If the colour of the layers does not change, then the substance belongs to nonionic surfactants.

Plant protection systems for peas were provided for: low (seed treatment by the drug Fundazol – 50% wettable powder (2 kg·t^{-1}); integrated, in addition to seed treatment by Fundazol, crops were treated by Bazagran herbicide – 48% aqueous solution (1.5 L·ha^{-1}) and Fusilade - 12.5% emulsion concentrate (0.8 L·ha^{-1}) in the phase of 3–4 pea leaves and Fastak insecticide – 10% emulsion concentrate (0.1 L·ha^{-1}) when pests was detected on the seedlings.

Statistical analysis. The least significant difference at $P < 0.05$). Statistical processing was performed by Microsoft Excel in combination with XLSTAT.

RESULTS AND DISCUSSION

The use of fertilizers and pesticides in intensive agriculture is a source of surfactants (SA) entering the agroecosystem, but the dependence of their content in the soil on the saturation of crop rotation with fertilizers and agrochemicals remains insufficiently studied. In the conditions of the model experiment, we studied the possibility of changing the amount of surfactants in the arable layer of soil under the influence of various agrochemical loads. To conduct a model experiment, the soil of the variant without fertilizers and the variant with maximum saturation of crop rotation with mineral fertilizers under different plant protection systems were used. In order to revive biochemical processes, the soil was composted for three days in optimal conditions (temperature – 25–28 °C with a soil moisture content of 60% of total moisture content) with subsequent determination of different groups of surfactants in it.

It was established that mineral fertilizers and pesticides increased the content of anionic surfactants (ASA) in the soil, and their complex application in a single unit enhanced the effect of each of the factors (Table 1). With the application of pesticides, an increase in the content of ASA in the soil by 22.7–27.9% was observed compared to the option without their application. Application of mineral fertilizers increased the ASA content in the soil by 5.2%. Complex application of fertilizers and pesticides increased the content of ASA in the soil by 86.7%.

In contrast to anionic surfactants, the content of nonionic surfactants increased by 30.3–40.5% due to the use of pesticides, but their decrease was observed when mineral fertilizers were used (-14.0%). The complex use of fertilizers and pesticides in one block neutralized the effect of each of the factors on the concentration of nonionic surfactants in the soil.

In contrast to the model experiment, where optimal hydrothermal conditions were artificially created, in field studies a number of factors act on the soil, therefore an analysis of the soil selected in the field after harvesting peas was additionally conducted. ASA content was determined in the three most contrasting options: absolute control without fertilizers and pesticides; $N_{129}P_{120}K_{148}$ + 10 t·ha^{-1} of manure; crop by-products (5 t ha^{-1} + $N_{17.5}$) + 10 t·ha^{-1} of manure. It was established that in field conditions the absolute values were significantly lower than under artificial conditions of the model experiment, although the main regularities were preserved (Figure 1).

Under the condition of growing crop rotation without pesticides and fertilizers or using only biological elements, the presence of ASA in the soil is close to their content in fallow (2 mg·kg^{-1} of soil), where agriculture has not been carried out for more than 20 years (fallow is a type of land (in agriculture) which represent lands which were in agricultural use, under arable land, but abandoned and not cultivated). The use of fertilizers and pesticides in grain-tilled crop rotation was

Table 1. The presence of SA in dark gray podzolized soil depending on the plant protection system and fertilizing options (model study)

Doses of fertilizers per 1 ha of crop rotation area	Plants protection system	Availability ASA in the soil		Availability NSA in the soil	
		mg kg^{-1} of soil	An increase from the pesticides use, %	mg kg^{-1} of soil	An increase from the pesticides use, %
Without fertilizers	I	174	+ 27.9	201	+ 40.5
Without fertilizers	II	136		143	
$N_{129}P_{120}K_{148}$	I	254	+ 22.7	159	+ 30.3
$N_{129}P_{120}K_{148}$	II	207		122	
An increase from fertilizers			+ 52.2		- 14.0
An increase from fertilizers			+ 86.7		+ 11.0

Note: I – integrated plant protection system; II – low plant protection system.

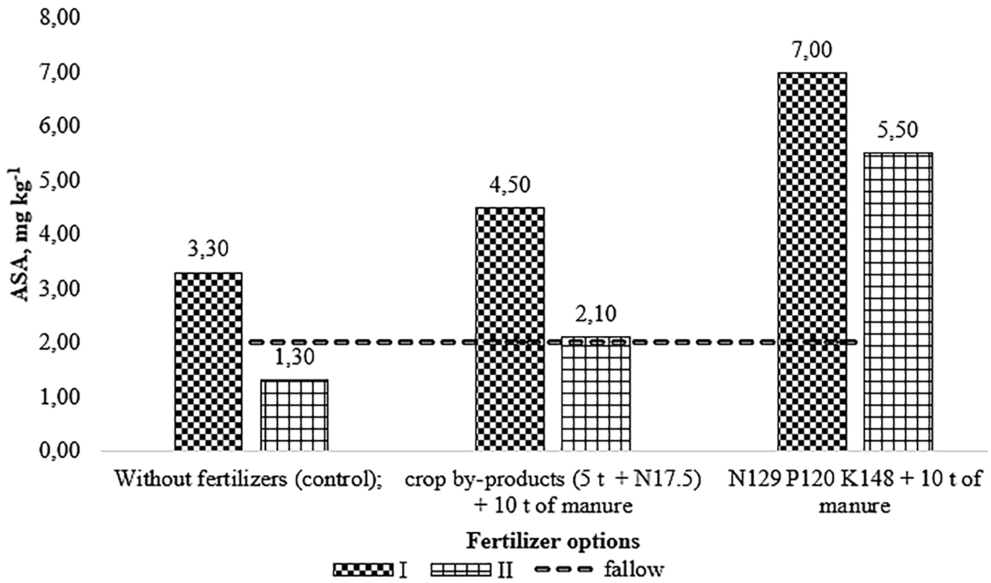

Figure 1. ASA content in dark gray podzolized soil depending on the plant protection system and fertilizing options; I – integrated plant protection system; II – low plant protection system

accompanied by an increase in ASA in the soil. Moreover, with an increase in the dose of mineral fertilizers and the intensity of pesticides use, the content of ASA increased.

In the soil samples taken after the test crop collection, along with surfactants determination, the nutrient regime of the soil was studied. It was established that the systematic application of organic and mineral fertilizers improved the nutritional regime of dark gray podzolized soil, as a result of which the content of biogenic elements in the top layer of the fertilized variants increased compared to the control. The amount of alkaline hydrolyzed nitrogen under the low protection system increased by 3.0–23.2 mg·kg^{-1} soil, mobile phosphorus (P_2O_5) by 14.0–144.0 mg, mobile potassium (K_2O) by 9.0–222.0 mg·kg^{-1} of soil, compared to the control. With the complex use of fertilizers and pesticides in one block, a trend of additional increase in mobile forms of nutrients in the soil was observed in the range from 0.7 to 7% of alkaline hydrolyzed nitrogen, from 2.0 to 15.0% of mobile phosphorus and from 3.0 to 5.0% of mobile potassium (Table 2).

The mechanism of the effect of surfactants is that their presence in the soil solution reduces the loss of moisture from the arable layer due to the formation of a convex meniscus in the soil capillaries, and facilitates the transition of nutrients into the soil solution (Prodanchuk & Mudriy, 2000). In addition, surfactants contribute to the retention of nutrients in fertilizers in a form available

Table 2. The influence of the systematic application of chemicals on the nutrient regime of dark gray podzolized soil, soil layer – 0–20 cm

Doses of fertilizers per 1 ha of crop rotation area	Alkaline hydrolyzed nitrogen, mg kg^{-1} of soil		Mobile phosphorus, P_2O_5, mg kg^{-1} of soil		Mobile potassium, K_2O, mg kg^{-1} of soil		Humus, %	
	I	II	I	II	I	II	I	II
Without fertilizers (control)	66.8 ^{g}	64.2 ^{g}	133 ^{g}	126 ^{g}	160 ^{g}	151 ^{g}	1.79 ^{d}	1.77 ^{d}
on background 10 t ha^{-1} of manure								
$N_{43}P_{40}K_{49}$	76.2 ^{d}	74.2 ^{e}	170 ^{e}	165 ^{e}	180 ^{d}	172 ^{d}	1.73 ^{e}	1.79 ^{d}
$N_{86}P_{80}K_{99}$	82.3 ^{b}	78.1 ^{c}	213 ^{c}	186 ^{c}	184 ^{c}	177 ^{c}	1.85 ^{b}	1.90 ^{b}
$N_{129}P_{120}K_{148}$	93.7 ^{a}	87.4 ^{a}	288 ^{b}	260 ^{b}	302 ^{b}	293 ^{b}	1.90 ^{a}	1.95 ^{a}
$N_{86}P_{80}K_{99}$	79.6 ^{c}	79.3 ^{b}	297 ^{a}	270 ^{a}	389 ^{a}	373 ^{a}	1.85 ^{b}	1.90 ^{b}
$N_{43}P_{40}K_{49}$ + by-products (5 t ha^{-1})	77.1 ^{e}	76.2 ^{d}	203 ^{d}	178 ^{d}	166 ^{e}	166 ^{e}	1.80 ^{c}	1.85 ^{c}
By-products (5 t ha^{-1} + $N_{17.5}$)	67.7 ^{f}	67.2 ^{f}	143 ^{f}	140 ^{f}	160 ^{f}	160 ^{f}	1.87 ^{ab}	1.85 ^{c}

Note: I – integrated plant protection system; II – low plant protection system. Means in columns with the different letter are highly significantly different according to the Fisher's test ($P \leq 0.05$).

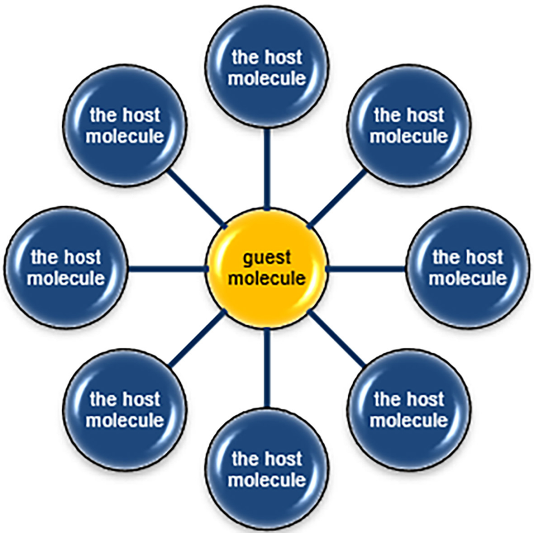

Figure 2. Scheme of the mechanism of surfactants action in the soil solution: surfactant – ('the host molecule'); a compound of a biogenic element ('guest molecule') (Prodanchuk & Mudriy, 2000)

to plants, preventing their irreplaceable fixation by soil components, since under the influence of surfactants, so-called «inclusion compounds» can occur. At the same time, the surfactant in the solution, like a «host molecule», wraps around a molecule of another substance («guest molecule») that entered the soil solution (Figure 2).

If the role of a «guest molecule» is a compound of a certain biogenic element, then this element, remaining available to the plant, will not be able to participate in chemical and physicochemical reactions with the soil, and therefore will not be permanently fixed in its solid phase. Only after surfactant shell destruction, the molecule of the biogenic element is released and can undergo processes of decay, synthesis, and inclusion in other compounds and complexes. Therefore, the presence of surfactants in the soil slows down the transition of nutrient elements into unavailable to plants forms, which contributes to the improvement of the nutritional conditions of agricultural plants. Cells of microorganisms can also act as "guest molecules", which is accompanied by a slowdown in their inclusion in biochemical processes due to partial isolation from the substrate. In the scientific literature, there is enough convincing evidence about the high adsorption capacity of ASA in various types of soils. However, it is noted that the rate and intensity of surfactant adsorption decrease with depth in the soil profile (Eliseev & Kucher, 1991).

When studying the effect of agrochemical load on the content of ASA in the profile of dark gray podzolized soil, we chose a fallow (an area that has not been cultivated for more than 20 years) as a control, and therefore chemical agents are not applied (Table 3).

Research has established that fertilizers and pesticides are a source of an additional influx of surface-active substances (surfactants), particularly

Table 3. ASA content in dark gray podzolized soil depending on the agrochemical load (layer 0–100 cm), mg kg^{-1} of soil

Depth of soil sample selection, sm	Saturation of crop rotation with mineral fertilizers		
	Fallow*	Crop by-products (5 t ha^{-1} + $N_{17.5}$) on background 10 t ha^{-1} of manure	$N_{129}P_{120}K_{148}$ on background 10 t ha^{-1} of manure
0–20	2.0	4.5	5.1
20–40	0.6	3.0	3.8
40–60	0.1	0.8	1.2
60–80	0.0	0.3	0.6
80–100	0.0	0.0	0.0

Note: Fallow - a type of land (in agriculture) or land that has been in agricultural use, under arable land, but abandoned and not cultivated.

their anionic forms, into the agro-ecosystem. The distribution of ASA amount in the soil profile is descending. On the fallow, the clear presence of ASA was established only in the soil layer of 0–40 cm. An increase in ASA content in the soil due to the use of agrochemicals and fertilizers is observed up to a depth of 60–80 cm. Using biological elements in agriculture significantly reduces the amount of these substances in the soil profile.

CONCLUSIONS

Surfactants play a crucial role in the soil ecosystem, and the use of agrochemicals such as fertilizers and pesticides can lead to their increased presence in soil. The research conducted showed that the use of pesticides in crop production led to an increase in the number of surfactants present in the soil, including both anionic (from 22.7% to 27.9%) and nonionic (from 30.3% to 40.5%) forms. When mineral fertilizers and pesticides were used together, there was a significant increase in the anionic form of surfactants, which reached 86.7%. This information highlights the impact of agricultural practices on soil health and the need to consider surfactants as a potential factor in soil management.

The presence of anionic surfactants (ASA) in the soil solution can contribute to improving the nutrient status of the topsoil and increase the concentration of mobile forms of biogenic elements throughout the soil profile. For example, it was found that the presence of ASA in the soil resulted in an increase of alkaline hydrolyzable nitrogen by 3–7%, mobile phosphorus by 3–15%, and mobile potassium by 3–6%.

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