

## Article

# Sewage Sludge Ash-Based Biofertilizers as a Circular Approach to Phosphorus: The Issue of Fe and Al in Soil and Wheat and Weed Plants

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**Abstract:** Sewage sludge management for fertilizer purposes can be a step in the circular phosphorus (P) economy. Using microbial solubilization in manufacturing fertilizers from recycled materials is an innovative approach with the potential to increase P compounds' bioavailability, and fertilizers from sewage sludge ash and P-solubilizing bacteria are promising products of this technology. In addition to P and a range of macronutrients, these fertilizers contain small amounts of micronutrients and potentially toxic elements. This paper discusses the effects of fertilizer on iron (Fe) and aluminum (Al) content in soil, test plants (spring or winter wheat; grain and straw), weeds and post-harvest residues, based on field experiments. Treatments with conventional P fertilizers (superphosphate, phosphorite) and without P fertilization provided references. The tested biofertilizers containing the *Bacillus megaterium* or *Acidithiobacillus ferrooxidans* strain had no effect on total Fe and Al content in the soil or on the concentration of these elements in plant biomass when applied at P doses up to 35.2 kg ha<sup>-1</sup>. Fe and Al levels in grain did not suggest a potential risk to consumers.

**Keywords:** nutrient recycling; secondary raw materials; waste management; phosphorus solubilizing microorganisms; heavy metals; *Triticum aestivum* L.



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## 1. Introduction

Sewage sludge is a problematic but so far unavoidable by-product of human civilization, generated during wastewater treatment. Information on the global production of sewage sludge is incomplete [1,2]. However, it is estimated at 45 million tons of dry matter per year, and Europe, East Asia, and North America are the main producers of sewage sludge globally [3]. It seems self-evident that the quantity of sludge produced will increase with the growing population and ongoing urbanization [4]. Following this, the need for the appropriate environmentally friendly management of this waste biomass will become more and more pressing. Different methods can be used to dispose of sewage sludge [5], and the choice of method mainly depends on the properties of the sludge [6], including organic and inorganic pollutant concentrations [7].

Increasing environmental awareness has resulted in legal limitations on sewage sludge disposal practices, especially in Europe. In 1986, substantial limitations for the application of sewage sludge in agriculture (as fertilizer/soil improver) were implemented [8]. In 1998, waste disposal in the sea/ocean was prohibited [9]. In 1999, limits were introduced for biodegradable wastes going into landfills [10].

In view of these legal aspects, sludge incineration is an increasingly widely accepted waste disposal option [11]. This method effectively reduces the volume of sewage sludge and eliminates pathogens and organic pollutants while producing heat and/or electricity [5].

Around 20% of sewage sludge produced in the US is incinerated, while in European Union member countries incineration levels are nearly 25% [11]. The rising amount of sludge being incinerated [2] necessitates the search for means to beneficially manage the incineration residue, i.e., sewage sludge ash (SSA).

SSA has been explored in recent years as a potential renewable raw material for phosphorus fertilizers and thus as a pathway to the re-inclusion of P in the food system [11–15].

Phosphorus is a crucial element for life (and food production). Still, the natural P cycle has been disrupted so much that humanity faces two linked problems: the dwindling supply of phosphate rock as a resource, and the overabundance of phosphate in water systems, leading to eutrophication [16]. Moreover, with a growing population, the United Nations' Sustainable Development Goal number two (zero hunger) will be difficult to achieve if the waste of phosphorus resources continues and the P cycle is not closed through its recovery and recycling [17]. These rationales call for increased efforts toward creating a circular economy for P [17], especially in Europe, for which phosphorus and phosphate rocks are among the most important critical raw materials [18]. Most European countries, including Poland, have to import phosphate rocks for mineral phosphate fertilizer production. Moreover, phosphate rock and the resulting mineral fertilizers contain relevant amounts of heavy metals (e.g., Cd and U) that transfer to the soil and could be a potential risk [19].

Although the idea of P recovery is fully theoretically justified, the practice of its implementation, however, faces many obstacles. Most importantly, nutrient prices in waste-based products are generally higher than in mineral fertilizers, and waste materials require additional post-treatment to increase nutrient concentration and availability [20]. Over the past years, numerous approaches to P recovery from SSA have been developed, such as thermo-chemical, acidic wet-chemical leaching, thermo-electric, and acidic wet-chemical extraction [14]. They are expected to enable the use of SSA for fertilizer production, mainly to enhance P bioavailability and reduce inorganic contaminant amounts [15].

An alternative for treating SSA with acids obtained from the chemical industry is using acids produced by microorganisms occurring in natural environments, e.g., soil [21], making P accessible for plants to absorb. Lowering environment pH by microbial production of organic acids (including acetic, citric, formic, fumaric, gluconic, lactic, malic, propionic, succinic, tartaric, valeric, and others) or release of protons is considered the principal mechanism for microbial solubilization of unavailable inorganic P compounds. Other mechanisms of mineral phosphate solubilization by microorganisms are the production of inorganic acids (such as sulfuric, nitric, and carbonic) and chelating substances [22]. A large number of autotrophic and heterotrophic soil microorganisms, aerobic and anaerobic, are able to solubilize various forms of insoluble phosphate compounds [23]. They can belong to diverse phyla, the most common of which are Proteobacteria, Firmicutes, Ascomycota, Euryarchaeota, Actinobacteria, Basidiomycota, Bacteroidetes, and Mucoromycota [24].

The mechanisms of microbial solubilization and mineralization of phosphorus compounds, the importance of these processes in agriculture, and the many strains of microorganisms capable of performing them have been described extensively in the existing literature [22,25,26]. Using microbial solubilization in the valorizing of secondary raw materials (including SSA) has also been discussed [21,27]. Innovative technology has been developed to produce biofertilizers following this concept and the favorable results of laboratory-scale studies. The major P source was SSA, and the biological agent activating this raw material was P-solubilizing bacteria [28]. The obtained bioproducts were evaluated under field conditions, and many of the findings have already been published [29,30]. Considering the chemical composition of SSA, in addition to P and a range of macronutrients, the biofertilizers contain small amounts of micronutrients and potentially toxic elements (PTEs). Therefore, introducing them into the soil–plant system can bring additional benefits (essential micronutrients) or contamination risks (toxic elements). The effect of SSA-based biofertilizers on the content of several PTEs (Cd, Pb, As, Cr, Ni, Cu, and Zn) in this system has been previously reported [31,32]. This paper addresses the issues of iron (Fe) and aluminum (Al). Fe and Al are usually abundant metal elements in SSA, which is associated

with Fe-based or Al-based salts for the precipitation of P from wastewater during the wastewater treatment process [33].

Iron (Fe) is an essential element for almost all living organisms, as it participates in a wide variety of metabolic processes, including oxygen transport, deoxyribonucleic acid (DNA) synthesis, and electron transport [34]. Fe deficiency remains a major threat to peoples' health around the world [35]. Thus, agronomic food bio-fortification (through crop fertilization) can be the key approach for mitigation of this micronutrient shortage [36]. However, excess accumulation of Fe can easily be toxic for plants [37] and animals [38]. Aluminum (Al) is not considered an essential element, and thus far no experimental evidence has been put forward for a biological role. In plants and other organisms, Al can have a beneficial or toxic effect, depending on factors such as metal concentration, the chemical form of Al, growth conditions, and plant species [39].

The aim of this paper is to assess the effect of biofertilizers from SSA and *Bacillus megaterium* (Firmicutes, Gram-positive) or *Acidithiobacillus ferrooxidans* (Proteobacteria, Gram-negative) bacteria on the content of Fe and Al in the soil, test plants (wheat grain and straw), weeds, and post-harvest residue biomass. It was hypothesized that the amounts of Fe and Al contributed by biofertilizers would not cause a threat to the soil–plant system in the agroecosystem.

## 2. Materials and Methods

In the years 2014–2016, five phosphorus biofertilizers produced from SSA were tested in field experiments conducted in Bałcyny (Poland, 53.60° N, 19.85° E). In some of them, the additional raw material was animal (poultry) bones or dried animal blood. Phosphorus-solubilizing bacteria from *Bacillus megaterium* (PCM 1855) or *Acidithiobacillus ferrooxidans* (F7-01) strains were used as biological activators of the phosphorus feedstock. General information about the studied biofertilizers is shown in Table 1, and their detailed chemical composition is provided in Table S1. The biofertilizers were produced by the New Chemical Syntheses Institute in Puławy (Poland) based on the formula developed by the Department of Advanced Material Technologies of the Wrocław University of Science and Technology (Poland). The production process is described elsewhere [40,41].

**Table 1.** Biofertilizers tested in the experiments.

Symbol	Raw Material	Bacteria	Form	P, % Mass	Fe, g kg <sup>-1</sup>	Al, g kg <sup>-1</sup>
A <sub>s</sub> B <sub>m</sub>	sewage sludge ash (SSA) <sup>1</sup>	<i>Bacillus megaterium</i> <sup>3</sup>	suspension	0.176	1.68	1.78
A <sub>g</sub> A <sub>f</sub>	SSA	<i>Acidithiobacillus ferrooxidans</i> <sup>4</sup>	granules	9.24	33.0	24.4
AB <sub>g</sub> A <sub>f</sub>	SSA + bones <sup>2</sup>	<i>Acidithiobacillus ferrooxidans</i>	granules	7.50	15.1	13.9
AB <sub>g</sub> B <sub>m</sub>	SSA + bones	<i>Bacillus megaterium</i>	granules	5.87	14.4	11.3
AH <sub>g</sub> B <sub>m</sub>	SSA + blood <sup>2</sup>	<i>Bacillus megaterium</i>	granules	9.55	29.0	25.5

<sup>1</sup> Obtained from Municipal Wastewater Treatment Plant 'Łyna' in Olsztyn, Poland; <sup>2</sup> obtained from the meat industry; <sup>3</sup> obtained from the Polish Collection of Microorganisms at the Institute of Immunology and Experimental Therapy of the Polish Academy of Sciences in Wrocław, Poland; <sup>4</sup> obtained from Professor Zygmunt Sadowski from Wrocław University of Science and Technology; the strain is autochthonous, isolated from the tailings impoundment "Iron Bridge", Poland [42].

In the experiments, common wheat (*Triticum aestivum* ssp. *vulgare* Mac Key), winter or spring, was used as a test plant. The referential P carriers for the tested biofertilizers were superphosphate Fosdar 40 (SP: 17.6% mass P; Grupa Azoty FOSFOR Sp. z o.o., Gdańsk, Poland), Syrian phosphorite (PR: 12.2–12.9% mass P, 0.79–1.61 g kg<sup>-1</sup> Fe, 0.80–1.69 g kg<sup>-1</sup> Al; Luvena, Luboń, Poland), an ash–water solution (A + H<sub>2</sub>O: 0.176% mass P, 1.68 g kg<sup>-1</sup> Fe, 1.77 g kg<sup>-1</sup> Al), and granular fertilizers from SSA + bones and SSA + blood without bacteria (AB<sub>g</sub>: 6.10% mass P, 15.4 g kg<sup>-1</sup> Fe, 12.8 g kg<sup>-1</sup> Al; AH<sub>g</sub>: 8.68% mass P, 26.9 g kg<sup>-1</sup> Fe, 23.7 g kg<sup>-1</sup> Al; New Chemical Syntheses Institute in Puławy, Poland). In each experiment, the reference treatment without phosphorus fertilization (No P) was also included. The

experimental details are summarized in Table 2, while basic agricultural data for the experiments are given in Table S2.

**Table 2.** Field experiments conducted—trial details.

Item	Experiment				
	I	II	III	IV	V
Year Test Plant	2014 spring wheat	2015 spring wheat	2015 winter wheat	2016 winter wheat	2016 spring wheat
Biofertilizers tested	$A_gBm$	$A_gBm$	$A_gAf$ $AB_gAf$	$AB_gBm$	$AH_gBm$
Reference treatments	no P SP PR A + H <sub>2</sub> O	no P SP PR	no P SP	no P SP $AB_g$	no P SP $AH_g$
P doses, kg ha <sup>-1</sup>	21	17.6 26.4 35.2	17.6 26.4 35.2	17.6 26.4 35.2	17.6 26.4 35.2
Plant Protection (PP)	PP–	PP– PP+	PP+	PP+	PP+
Number of treatment combinations	5	20	10	10	10
Experimental design	randomized block	parallel strip	randomized block	randomized block	randomized block
Number of replications	4	4	4	4	4
Number of plots	20	80	40	40	40

PP–: no plant protection treatments; PP+: full plant protection (see Table S2).

Wheat was grown on Luvisols [43] formed from sandy clay loam or sandy loam. The soil pH<sub>KCl</sub> of the arable layer (0–30 cm) was 5.23–6.28, and the total C, N, P, K, Mg, and P contents were 7.15–8.90 g kg<sup>-1</sup>, 1.09–1.42 g kg<sup>-1</sup>, 0.43–0.61 g kg<sup>-1</sup>, 2.90–3.30 g kg<sup>-1</sup>, 2.01–2.25 g kg<sup>-1</sup>, and 0.433–0.610 g kg<sup>-1</sup>, respectively (Table S3). Precipitation and thermal conditions in the growing seasons differed from those typical of the region, being too dry for wheat in experiments I–III, but too wet for this species in experiments IV and V (Table S4).

Soil samples for chemical analyses were taken from the 0–30 cm soil layer twice, i.e., before the start of the experiment and after wheat harvest. The samples of wheat straw (i.e., wheat stems with leaves) and weeds were collected shortly before wheat harvest, while wheat grain and the post-harvest residues (wheat roots, bottom stem segments, weed remnants) were sampled after harvest.

Soil total C and N contents were determined using a Vario Macro Cube Elementar (C, H, N) analyzer (Elementar Analysensysteme, Langensfeld, Germany) and D-phenylalanine (C = 65.44%; N = 8.48%) as a standard solution. The contents of total P, K, Mg, Fe, and Al in the soil and Fe and Al contents in plant material were determined using an inductively coupled plasma-optical emission spectrometer (ICP–OES with a pneumatic nebulizer with an axial view—iCAP Duo Thermo Scientific, Waltham, MA, USA). The levels of detection (LOD) for P, K, Mg, Fe, and Al for the soil material were 3.59, 2.55, 1.17, 0.72, and 2.34 mg kg<sup>-1</sup>, respectively, and for the plant material, the detection levels for Fe and Al were: 0.040 and 0.015 mg kg<sup>-1</sup>, respectively. More detailed descriptions of soil and plant sampling, preparation, and chemical analyses are provided in the Supporting Information.

The raw data were submitted to an analysis of variance (ANOVA) or the alternative Kruskal–Wallis test (if ANOVA assumptions were not met). The normality of variable distribution was checked using the Shapiro–Wilk W-test, and the homogeneity of variance was checked using Levene’s test. Finally, the differences between objects were assessed using Duncan’s or multiple comparison test. The calculations were performed using Statistica 13.3 software [44].

### 3. Results and Discussion

#### 3.1. Iron (Fe)

The level of total Fe content in the soil in the conducted experiments (Table 3) was within the region-specific range (4.0–25.8 g kg<sup>-1</sup>) and closer to the national average value (9.0 g kg<sup>-1</sup>) than the median (6.6 g kg<sup>-1</sup>) [45]. It also corresponded with the average natural Fe content in the parent rocks of soils in Poland (geochemical background—12.9 g kg<sup>-1</sup>) [46]. The parent material primarily determines the total amount of iron in the soil [47]; however, Fe can also enter the soil from anthropogenic sources, including fertilizers or soil improvers derived from sewage sludge [48].

**Table 3.** Total Fe content in soil and plant biomass (average ± standard error).

Experiment	P-Fertilizer	P-Dose, kg ha <sup>-1</sup>	Plant Protection	Soil, g kg <sup>-1</sup> DM		Plant Biomass, mg kg <sup>-1</sup> DM					
				Start	End	Wheat Grain	Wheat Straw	Weeds	Post-Harvest Residues		
I	No P	0	PP–	11.2 ± 0.8	10.8 ± 0.5	39.4 ± 1.4	54 ± 5	181 ± 24	2441 ± 155		
	SP	21		11.5 ± 0.6	11.5 ± 0.8	39.4 ± 1.4	54 ± 2	157 ± 19	2519 ± 254		
	PR	21		10.9 ± 0.3	11.1 ± 0.6	39.4 ± 0.8	61 ± 6	209 ± 29	2370 ± 176		
	A + H <sub>2</sub> O	21		11.4 ± 0.8	11.3 ± 0.5	39.2 ± 1.1	53 ± 6	192 ± 15	2699 ± 217		
	A <sub>s</sub> B <sub>m</sub>	21		11.3 ± 1.0	11.3 ± 0.7	41.1 ± 1.8	51 ± 3	198 ± 21	2603 ± 162		
II	No P	0	PP–	11.1 ± 0.4	9.2 ± 0.9↓ <sup>1</sup>	62.1 ± 8.4	102 ± 8	442 ± 125	2080 ± 394		
	SP	17.6		11.0 ± 0.3	9.1 ± 0.9↓	51.1 ± 2.1	98 ± 14	418 ± 69	1798 ± 286		
		26.4		10.7 ± 0.6	9.2 ± 0.9↓	55.5 ± 7.9	100 ± 3	425 ± 133	1761 ± 204		
		35.2		11.2 ± 0.3	9.1 ± 0.8↓	54.0 ± 6.8	94 ± 11	514 ± 149	1970 ± 222		
	PR	17.6		10.6 ± 0.4	9.2 ± 0.9↓	53.6 ± 7.3	118 ± 14	640 ± 188	1846 ± 176		
		26.4		11.0 ± 0.5	9.2 ± 0.5↓	62.7 ± 8.3	103 ± 20	495 ± 114	1926 ± 229		
		35.2		10.5 ± 0.5	9.1 ± 0.8↓	48.0 ± 5.8	100 ± 14	492 ± 86	1638 ± 347		
	A <sub>s</sub> B <sub>m</sub>	17.6		11.3 ± 0.4	9.2 ± 0.9↓	57.5 ± 11.9	90 ± 5	416 ± 96	1740 ± 269		
		26.4		10.7 ± 0.5	9.1 ± 0.9↓	57.6 ± 7.4	101 ± 19	669 ± 192	2143 ± 296		
		35.2		11.1 ± 0.4	9.3 ± 1.1↓	53.9 ± 7.3	99 ± 15	724 ± 274	1691 ± 234		
		average			10.9 ± 0.1	9.2 ± 0.2↓	55.6 ± 2.3	101 ± 4	524 ± 46 b <sup>2</sup>	1859 ± 80 b	
	No P	0		PP+	11.5 ± 0.7	9.3 ± 0.7↓	57.9 ± 4.2	92 ± 12	638 ± 29	2284 ± 520	
	SP	17.6			11.1 ± 0.6	9.3 ± 0.6↓	51.3 ± 7.7	92 ± 6	603 ± 159	2253 ± 159	
		26.4			10.9 ± 0.3	9.7 ± 0.5↓	60.8 ± 11.4	99 ± 13	666 ± 99	2154 ± 262	
		35.2			11.5 ± 0.5	9.5 ± 0.5↓	55.5 ± 9.4	106 ± 15	633 ± 162	2177 ± 88	
	PR	17.6			11.0 ± 0.5	9.0 ± 0.4↓	51.6 ± 6.1	86 ± 8	662 ± 3	2655 ± 494	
		26.4			10.7 ± 0.2	9.2 ± 0.5↓	56.2 ± 1.2	93 ± 9	608 ± 124	2502 ± 368	
		35.2			11.0 ± 0.3	9.0 ± 0.6↓	63.7 ± 9.6	84 ± 5	597 ± 171	2291 ± 330	
	A <sub>s</sub> B <sub>m</sub>	17.6			10.7 ± 0.3	8.6 ± 0.2↓	59.1 ± 2.7	105 ± 10	553 ± 131	2422 ± 221	
		26.4			10.7 ± 0.2	9.1 ± 0.3↓	59.6 ± 0.4	84 ± 10	917 ± 164	2201 ± 186	
	35.2	10.2 ± 0.1	9.3 ± 0.3↓		57.9 ± 7.0	103 ± 11	797 ± 34	2408 ± 315			
	average		10.9 ± 0.1		9.2 ± 0.1↓	57.0 ± 2.1	95 ± 3	667 ± 38 a	2335 ± 93 a		
III	No P	0	PP+		11.9 ± 0.5	10.8 ± 0.6↓	69.0 ± 7.8	80 ± 14	572 ± 102	3398 ± 489	
	SP	17.6			11.6 ± 0.6	10.1 ± 0.5↓	53.3 ± 3.3	69 ± 13	527 ± 97	3171 ± 120	
		26.4			11.8 ± 0.8	10.4 ± 0.6↓	52.4 ± 3.8	79 ± 12	493 ± 91	3546 ± 507	
		35.2			11.5 ± 0.5	10.6 ± 0.5↓	61.8 ± 9.3	72 ± 9	503 ± 112	3493 ± 351	
	A <sub>g</sub> A <sub>f</sub>	17.6			11.3 ± 0.9	10.2 ± 0.4↓	64.8 ± 5.5	79 ± 9	528 ± 103	3238 ± 343	
		26.4			11.3 ± 0.5	10.2 ± 0.4↓	53.0 ± 3.8	79 ± 5	632 ± 116	3628 ± 215	
		35.2			11.5 ± 0.8	10.4 ± 0.5↓	69.1 ± 9.9	71 ± 7	577 ± 52	3187 ± 372	
	AB <sub>g</sub> A <sub>f</sub>	17.6			11.7 ± 0.5	10.4 ± 0.5↓	59.4 ± 8.9	85 ± 8	596 ± 26	2975 ± 190	
		26.4			11.6 ± 0.7	10.5 ± 0.5↓	55.6 ± 4.6	89 ± 13	619 ± 93	3633 ± 475	
		35.2		11.3 ± 0.4	10.6 ± 0.5↓	57.6 ± 5.2	66 ± 9	673 ± 74	3230 ± 440		
	IV	No P		0	PP+	12.1 ± 0.6	10.1 ± 0.4↓	31.4 ± 1.1	105 ± 9	386 ± 66	1737 ± 285
		SP		17.6		11.4 ± 0.3	10.7 ± 0.7↓	31.9 ± 0.9	104 ± 18	417 ± 47	1948 ± 274
		26.4	12.1 ± 0.6	10.4 ± 0.3↓		31.3 ± 1.1	118 ± 19	392 ± 82	2273 ± 389		
		35.2	11.6 ± 0.4	10.2 ± 0.4↓		29.2 ± 0.3	117 ± 23	452 ± 182	1757 ± 243		
AB <sub>g</sub>		17.6	11.7 ± 0.8	10.8 ± 0.5↓		35.3 ± 1.0	126 ± 12	457 ± 76	2138 ± 204		
		26.4	11.8 ± 0.5	10.6 ± 0.3↓		30.9 ± 0.4	118 ± 12	565 ± 73	2172 ± 114		
		35.2	12.0 ± 0.8	10.7 ± 0.6↓		32.1 ± 0.9	135 ± 26	410 ± 14	1908 ± 185		
AB <sub>g</sub> B <sub>m</sub>		17.6	11.6 ± 0.5	10.7 ± 0.6↓		34.5 ± 1.9	114 ± 24	395 ± 90	2118 ± 153		
		26.4	11.5 ± 0.4	11.1 ± 0.7↓		33.0 ± 2.3	115 ± 26	545 ± 32	2055 ± 285		
		35.2	11.8 ± 0.8	10.9 ± 0.5↓		37.4 ± 6.0	100 ± 19	482 ± 137	2151 ± 355		

Table 3. Cont.

Experiment	P-Fertilizer	P-Dose, kg ha <sup>-1</sup>	Plant Protection	Soil, g kg <sup>-1</sup> DM		Plant Biomass, mg kg <sup>-1</sup> DM					
				Start	End	Wheat Grain	Wheat Straw	Weeds	Post-Harvest Residues		
V	No P	0	PP+	11.0 ± 0.9	10.8 ± 0.7	41.4 ± 1.6	101 ± 19	552 ± 85	2071 ± 62		
		SP		17.6	10.4 ± 0.5	10.5 ± 0.3	37.7 ± 1.1	97 ± 28	721 ± 107	2646 ± 190	
		26.4		10.9 ± 0.6	11.0 ± 0.5	38.2 ± 2.3	94 ± 21	581 ± 26	1969 ± 303		
	AH <sub>g</sub>	35.2		11.2 ± 0.7	10.8 ± 0.6	38.4 ± 1.1	136 ± 25	781 ± 176	2278 ± 454		
		17.6		10.6 ± 0.8	11.0 ± 0.4	46.6 ± 4.4	122 ± 30	613 ± 70	2121 ± 489		
		26.4		10.6 ± 0.5	10.6 ± 0.4	40.9 ± 2.1	129 ± 33	493 ± 94	2595 ± 423		
	AH <sub>g</sub> Bm	35.2		11.1 ± 0.6	10.8 ± 0.4	45.6 ± 7.7	112 ± 24	812 ± 342	2101 ± 333		
		17.6		10.3 ± 0.5	10.6 ± 0.5	37.6 ± 2.0	136 ± 25	630 ± 102	2498 ± 435		
		26.4		10.6 ± 0.8	10.6 ± 0.5	43.4 ± 2.2	129 ± 37	715 ± 172	1747 ± 182		
				35.2		10.4 ± 0.6	10.7 ± 0.5	38.2 ± 1.3	115 ± 19	538 ± 122	1763 ± 141

<sup>1</sup> arrows (↓) indicate significant decrease in relation to the starting state; <sup>2</sup> different letters indicate significant differences at  $p < 0.05$ ; no letters—no significant differences for plant protection treatments or for P-fertilization treatments.

In the present study, in none of the experiments conducted did P fertilization in the form of conventional and SSA-based fertilizers, at different P rates, differentiate the total Fe content in the soil. Such an effect is not surprising. Previous reports by other authors show that the application of fertilizers or soil amendments containing Fe even for a long period does not bring a significant change in the total Fe content in the soil but rather affects the amount of its mobile/available forms [48–50]. Assuming that the weight of dry soil from a 0–30 cm deep layer and an area of 1 ha equals 4500 t, the amount of Fe added with conventional and SSA-based fertilizers ranged from 0.025 mg kg<sup>-1</sup> of soil (applied with PR at the P dose of 17.6 kg ha<sup>-1</sup>) to 7.46 mg kg<sup>-1</sup> of soil (applied with A<sub>s</sub>Bm at the P dose of 35.2 kg ha<sup>-1</sup>). Some amounts of Fe are also introduced with SP (Fe content not defined in the commercial label). Nevertheless, these amounts were low compared with the high background total soil Fe content and thus negligible for statistical analysis. Furthermore, the level of total soil Fe after crop harvest was strongly influenced by plant uptake (wheat and weeds). Leaching of Fe from the soil could not be excluded either [48,51], but this was not the subject of the present study.

A decrease in the total Fe content in the soil compared to the initial state was observed after wheat harvest in experiments II–IV, i.e., where the soil pH was 5.23–5.51. In experiments I and V, where soil pH exceeded 6.0, no significant change in the total Fe content over time was found. This situation is explained by the dependence of the solubility of Fe compounds on soil pH. It is well known that the concentration of Fe in the soil solution decreases sharply as the soil pH increases [52]. Thus, in acidic soils (experiments II–IV), a higher abundance of mobile Fe (both from soil reserves and from fertilizers) resulted in a depletion of the total pool of this element by plant uptake or leaching deep into the soil profile.

None of the conducted experiments confirmed an increase in Fe content in plant biomass (wheat grains and straw, weeds, post-harvest residues) under P fertilization compared to the control. No differences were found between the effects of conventional and SSA-based fertilizers, regardless of the P dose. Such a result should be explained by the fact that Fe in SSA is present in the form of hematite [53], which is poorly soluble, and the potential of P-solubilizing bacteria contained in biofertilizers in making Fe available to plants (lowering pH by acid production [28], production of Fe chelating siderophores [54]) did not become apparent under the given experimental conditions. For comparison, in the study by Płaza et al. [55], an application of the *Bacillus megaterium* var. *phosphaticum* bacteria (dose of biological preparation: 1 L ha<sup>-1</sup>) increased Fe concentration in spring wheat grain and straw.

The Fe concentration in the studied plant biomass elements was arranged according to the following pattern: wheat grain < wheat straw < weed aboveground biomass < post-harvest residues. The presented results concerning Fe levels in wheat grain and straw were close to those obtained by other authors from Poland [55–58]. According to Kabata-Pendias and Pendias [59], the concentration of Fe in cereal grains fluctuates but rarely exceeds

100 mg kg<sup>-1</sup> DM. In national or international regulations, there are no set limits for Fe content in cereal grains for food and feed purposes [60–64]. Moreover, Fe content in wheat grain is generally considered low, and attempts have been made worldwide to increase it through agronomic and genetic biofortification [65]. In the present study, Fe content in wheat grain obtained in experiments II and III was close to, and sometimes even higher than, the target Fe content in wheat grain assumed in the HarvestPlus program (59 mg kg<sup>-1</sup> DM) [66]. However, this was not related to the application of the tested biofertilizers, unlike in the study by Plaza et al. [55], which reported an increase in Fe content in spring wheat grain from 34.2 to 68.6 mg kg<sup>-1</sup> after *Bacillus megaterium* application. Fe content in wheat straw oscillated around the value reported by Kirkby [67] as the average concentration of Fe in plant shoot dry matter sufficient for adequate growth (100 mg kg<sup>-1</sup>) and fell within the range of contents considered by Schulte [52] to be sufficient for wheat growth (20–250 mg kg<sup>-1</sup>; top leaves, boot stage).

The Fe content of aboveground weed biomass in experiments II–V seems to be high compared to the values presented by Liwski [68] and Głowacka [69]. It corresponds, however, with the ranges of Fe content reported by Rogóż and Niemiec [70] for different weed species growing on soils of similar pH and total Fe content to the present study. The Fe content of the biomass of post-harvest residue in all experiments was very high compared to that of wheat grain and straw. This was primarily due to the Fe levels in plant roots, which can contain up to several times more Fe than their aboveground parts [71,72]. There are no precisely defined critical levels of iron deficiency or excess/toxicity in plants. According to Kabata-Pendias [73], injured leaves indicated an accumulation of Fe above 1000 mg kg<sup>-1</sup>, while no information on toxic Fe levels in plant roots was found. With contemporary knowledge, plant response to Fe deficiency/toxicity is highly dependent on plant species, soil, and nutritional and climatic factors [73,74], and plants have developed effective mechanisms to maintain Fe homeostasis [75]. After absorption by roots, Fe first accumulates at the basal part of the shoot (discrimination center), and then to translocated shoots and to parts with a high Fe demand. Fe is readily oxidized and precipitated in the apoplast of both roots and shoots. The results of the present study suggest that much of the Fe uptake was retained in the roots.

In experiment II, plant protection effects on Fe content of weed and post-harvest residue biomass were found. In the case of weeds, the differences (PP– vs. PP+) are probably due to a change in the species structure of the weed community under the influence of herbicides, as different weed species can accumulate different amounts of Fe [69,70]. In turn, the differences between Fe contents in post-harvest residues under PP+ versus PP– conditions were mainly due to the ratio of roots to aboveground plant parts in the residue biomass. Under PP+ conditions, roots (mainly wheat) had higher biomass and, at the same time, a higher contribution to the total biomass of the residues, and these can store larger amounts of Fe [70,72].

### 3.2. Aluminum (Al)

The contents of total Al in soils in the present study (Table 4) were quite high considering the ranges established for Poland (range 1.2–18.1 g kg<sup>-1</sup>; average 5.7 g kg<sup>-1</sup>; median 4.5 g kg<sup>-1</sup>) and the region (2.9–11.2 g kg<sup>-1</sup>) within the framework of national monitoring [45]. These values seemed more optimistic compared to the range commonly cited in the literature, i.e., 10–250 g kg<sup>-1</sup> [76,77]. Total Al content in soils depends on both natural (parent rock, soil type) and anthropogenic sources (including P fertilizers); however, for agriculture, the most important are active Al forms [39,76,77]. National and EU regulations do not set limits for Al content in soils or fertilizers [78–80]. However, the use of sewage sludge, which typically contains Al compounds, is limited to soils whose pH is above 5.6 [81]. Some authors have called for bioavailable Al to be added to lists of standardized elements and for monitoring bioavailable Al content [82].

**Table 4.** Total Al content in soil and plant biomass (average  $\pm$  standard error).

Experiment	P-Fertilizer	P-dose, kg ha <sup>-1</sup>	Plant Protection	Soil, g kg <sup>-1</sup> DM		Plant Biomass, mg kg <sup>-1</sup> DM					
				Start	End	Wheat Grain	Wheat Straw	Weeds	Post-Harvest Residues		
I	No P	0	PP-	11.6 $\pm$ 0.9	10.8 $\pm$ 0.4	8.98 $\pm$ 0.81	63.1 $\pm$ 4.4	226 $\pm$ 47	3159 $\pm$ 309		
	SP	21		11.2 $\pm$ 0.5	11.8 $\pm$ 1.0	8.18 $\pm$ 0.68	59.0 $\pm$ 2.7	160 $\pm$ 20	3267 $\pm$ 449		
	PR	21		11.1 $\pm$ 0.2	11.3 $\pm$ 0.5	10.26 $\pm$ 1.59	68.8 $\pm$ 9.3	257 $\pm$ 34	3020 $\pm$ 298		
	A + H <sub>2</sub> O	21		11.4 $\pm$ 0.8	11.3 $\pm$ 0.4	8.83 $\pm$ 0.25	61.9 $\pm$ 9.8	411 $\pm$ 113	3444 $\pm$ 255		
	A <sub>s</sub> B <sub>m</sub>	21		11.9 $\pm$ 1.1	11.6 $\pm$ 0.8	8.77 $\pm$ 0.82	55.3 $\pm$ 1.1	218 $\pm$ 18	3255 $\pm$ 257		
II	No P	0	PP-	12.1 $\pm$ 0.5	10.0 $\pm$ 1.3 <sup>1</sup>	<LOD	121 $\pm$ 11	464 $\pm$ 136	2177 $\pm$ 443		
	SP	17.6		11.3 $\pm$ 0.7	9.9 $\pm$ 1.2 <sup>1</sup>	<LOD	132 $\pm$ 26	434 $\pm$ 66	1578 $\pm$ 303		
		26.4		11.2 $\pm$ 0.6	9.5 $\pm$ 0.8 <sup>1</sup>	<LOD	93 $\pm$ 9	416 $\pm$ 148	1615 $\pm$ 162		
		35.2		12.0 $\pm$ 0.4	9.8 $\pm$ 1.1 <sup>1</sup>	<LOD	91 $\pm$ 10	535 $\pm$ 167	1978 $\pm$ 251		
	PR	17.6		11.7 $\pm$ 0.6	9.5 $\pm$ 0.8 <sup>1</sup>	<LOD	150 $\pm$ 41	614 $\pm$ 202	1816 $\pm$ 178		
		26.4		11.6 $\pm$ 0.8	10.0 $\pm$ 1.0 <sup>1</sup>	<LOD	178 $\pm$ 13	530 $\pm$ 131	1895 $\pm$ 160		
		35.2		11.5 $\pm$ 0.7	9.5 $\pm$ 0.8 <sup>1</sup>	<LOD	103 $\pm$ 17	499 $\pm$ 86	1546 $\pm$ 265		
	A <sub>s</sub> B <sub>m</sub>	17.6		12.5 $\pm$ 0.5	8.9 $\pm$ 0.2 <sup>1</sup>	<LOD	100 $\pm$ 13	425 $\pm$ 93	1933 $\pm$ 378		
		26.4		11.1 $\pm$ 0.8	9.7 $\pm$ 1.0 <sup>1</sup>	<LOD	106 $\pm$ 15	705 $\pm$ 209	2628 $\pm$ 453		
		35.2		12.1 $\pm$ 0.6	9.8 $\pm$ 1.1 <sup>1</sup>	<LOD	110 $\pm$ 14	559 $\pm$ 185	1552 $\pm$ 181		
		average			11.7 $\pm$ 0.2	9.7 $\pm$ 0.3 <sup>1</sup>	<LOD	118 $\pm$ 7	518 $\pm$ 44 b <sup>2</sup>	1872 $\pm$ 97	
	No P	0		PP+	12.4 $\pm$ 0.7	10.3 $\pm$ 1.0 <sup>1</sup>	<LOD	95 $\pm$ 11	744 $\pm$ 29	1926 $\pm$ 626	
	SP	17.6			11.9 $\pm$ 0.6	10.4 $\pm$ 0.9 <sup>1</sup>	<LOD	119 $\pm$ 35	653 $\pm$ 161	1786 $\pm$ 172	
		26.4			11.5 $\pm$ 0.3	11.1 $\pm$ 0.7 <sup>1</sup>	<LOD	99 $\pm$ 23	670 $\pm$ 105	1397 $\pm$ 82	
		35.2			12.2 $\pm$ 0.5	10.8 $\pm$ 0.7 <sup>1</sup>	<LOD	119 $\pm$ 19	708 $\pm$ 200	1719 $\pm$ 230	
	PR	17.6			11.9 $\pm$ 0.4	10.0 $\pm$ 0.6 <sup>1</sup>	<LOD	131 $\pm$ 52	756 $\pm$ 17	2117 $\pm$ 409	
		26.4			11.4 $\pm$ 0.3	10.3 $\pm$ 0.8 <sup>1</sup>	<LOD	99 $\pm$ 3	667 $\pm$ 138	2592 $\pm$ 234	
		35.2			11.9 $\pm$ 0.3	9.9 $\pm$ 0.9 <sup>1</sup>	<LOD	91 $\pm$ 10	655 $\pm$ 199	1818 $\pm$ 353	
	A <sub>s</sub> B <sub>m</sub>	17.6			11.5 $\pm$ 0.2	9.2 $\pm$ 0.4 <sup>1</sup>	<LOD	99 $\pm$ 11	601 $\pm$ 133	2446 $\pm$ 666	
		26.4			11.4 $\pm$ 0.2	9.8 $\pm$ 0.7 <sup>1</sup>	<LOD	90 $\pm$ 12	985 $\pm$ 160	1691 $\pm$ 221	
	35.2	11.0 $\pm$ 0.1	10.2 $\pm$ 0.6 <sup>1</sup>		<LOD	107 $\pm$ 11	907 $\pm$ 64	2077 $\pm$ 267			
	average		11.7 $\pm$ 0.1		10.2 $\pm$ 0.2 <sup>1</sup>	<LOD	105 $\pm$ 7	735 $\pm$ 42 a	1957 $\pm$ 117		
III	No P	0	PP+		13.5 $\pm$ 0.7	12.4 $\pm$ 0.6 <sup>1</sup>	<LOD	121 $\pm$ 20	430 $\pm$ 97	3155 $\pm$ 411	
	SP	17.6			12.8 $\pm$ 0.7	11.7 $\pm$ 1.0 <sup>1</sup>	<LOD	99 $\pm$ 29	423 $\pm$ 92	2950 $\pm$ 238	
		26.4			13.2 $\pm$ 0.9	11.9 $\pm$ 1.3 <sup>1</sup>	<LOD	102 $\pm$ 17	376 $\pm$ 92	3143 $\pm$ 510	
		35.2			12.6 $\pm$ 0.6	12.1 $\pm$ 0.5 <sup>1</sup>	<LOD	97 $\pm$ 15	495 $\pm$ 160	3225 $\pm$ 302	
	A <sub>g</sub> A <sub>f</sub>	17.6			12.7 $\pm$ 1.1	12.2 $\pm$ 0.5 <sup>1</sup>	<LOD	106 $\pm$ 11	530 $\pm$ 145	3080 $\pm$ 319	
		26.4			12.8 $\pm$ 0.6	12.0 $\pm$ 0.8 <sup>1</sup>	<LOD	125 $\pm$ 15	537 $\pm$ 113	3236 $\pm$ 196	
		35.2			12.8 $\pm$ 1.0	12.1 $\pm$ 0.6 <sup>1</sup>	<LOD	96 $\pm$ 14	562 $\pm$ 108	2797 $\pm$ 297	
	AB <sub>g</sub> A <sub>f</sub>	17.6			13.1 $\pm$ 0.7	12.5 $\pm$ 0.5 <sup>1</sup>	<LOD	119 $\pm$ 10	492 $\pm$ 25	3040 $\pm$ 153	
		26.4			13.1 $\pm$ 0.8	12.5 $\pm$ 0.8 <sup>1</sup>	<LOD	117 $\pm$ 22	474 $\pm$ 72	3310 $\pm$ 417	
		35.2		12.6 $\pm$ 0.7	12.2 $\pm$ 0.5 <sup>1</sup>	<LOD	93 $\pm$ 9	596 $\pm$ 59	3141 $\pm$ 404		
	IV	No P		0	PP+	13.4 $\pm$ 1.0	11.0 $\pm$ 0.3 <sup>1</sup>	2.13 $\pm$ 1.54	137 $\pm$ 19	457 $\pm$ 86	1256 $\pm$ 212
		SP		17.6		13.2 $\pm$ 0.4	11.8 $\pm$ 0.9 <sup>1</sup>	1.85 $\pm$ 0.81	131 $\pm$ 31	528 $\pm$ 82	1427 $\pm$ 220
		26.4	13.6 $\pm$ 0.6	11.9 $\pm$ 0.3 <sup>1</sup>		2.11 $\pm$ 0.78	165 $\pm$ 37	483 $\pm$ 114	1362 $\pm$ 204		
		35.2	13.3 $\pm$ 0.5	11.5 $\pm$ 0.6 <sup>1</sup>		2.89 $\pm$ 1.10	149 $\pm$ 27	405 $\pm$ 95	1414 $\pm$ 100		
AB <sub>g</sub>		17.6	13.3 $\pm$ 0.9	12.0 $\pm$ 0.6 <sup>1</sup>		2.21 $\pm$ 0.93	156 $\pm$ 17	489 $\pm$ 146	1662 $\pm$ 116		
		26.4	13.3 $\pm$ 0.7	11.6 $\pm$ 0.4 <sup>1</sup>		1.72 $\pm$ 0.84	162 $\pm$ 29	676 $\pm$ 65	1435 $\pm$ 124		
		35.2	13.7 $\pm$ 1.0	11.9 $\pm$ 0.7 <sup>1</sup>		4.09 $\pm$ 1.65	169 $\pm$ 30	514 $\pm$ 26	1402 $\pm$ 131		
AB <sub>g</sub> B <sub>m</sub>		17.6	12.6 $\pm$ 1.0	11.6 $\pm$ 0.9 <sup>1</sup>		5.80 $\pm$ 1.31	142 $\pm$ 17	417 $\pm$ 105	1459 $\pm$ 108		
		26.4	12.9 $\pm$ 0.3	12.7 $\pm$ 0.7 <sup>1</sup>		1.52 $\pm$ 0.17	134 $\pm$ 26	662 $\pm$ 42	1409 $\pm$ 236		
		35.2	13.5 $\pm$ 1.2	12.3 $\pm$ 0.8 <sup>1</sup>		2.62 $\pm$ 0.42	179 $\pm$ 30	488 $\pm$ 103	1411 $\pm$ 193		
V		No P	0	PP+		12.5 $\pm$ 1.0	12.0 $\pm$ 0.9	2.36 $\pm$ 1.55	139 $\pm$ 25	785 $\pm$ 146	2445 $\pm$ 43
		SP	17.6			12.1 $\pm$ 0.3	12.0 $\pm$ 0.5	4.31 $\pm$ 1.04	159 $\pm$ 36	964 $\pm$ 162	3013 $\pm$ 200
		26.4	12.2 $\pm$ 0.9		12.2 $\pm$ 0.7	3.33 $\pm$ 0.74	125 $\pm$ 31	756 $\pm$ 18	2232 $\pm$ 362		
		35.2	12.7 $\pm$ 0.7		12.0 $\pm$ 0.4	2.71 $\pm$ 1.42	181 $\pm$ 41	911 $\pm$ 183	2404 $\pm$ 443		
	AH <sub>g</sub>	17.6	12.9 $\pm$ 0.5		12.2 $\pm$ 0.3	3.18 $\pm$ 1.01	166 $\pm$ 47	824 $\pm$ 140	2337 $\pm$ 344		
		26.4	11.7 $\pm$ 0.5		12.0 $\pm$ 0.5	2.74 $\pm$ 0.84	184 $\pm$ 47	707 $\pm$ 145	3049 $\pm$ 450		
		35.2	12.1 $\pm$ 0.8		12.0 $\pm$ 0.5	1.40 $\pm$ 0.82	152 $\pm$ 40	902 $\pm$ 251	2392 $\pm$ 327		
	AH <sub>g</sub> B <sub>m</sub>	17.6	12.6 $\pm$ 0.0		11.9 $\pm$ 0.6	1.35 $\pm$ 0.77	186 $\pm$ 36	871 $\pm$ 166	2797 $\pm$ 437		
		26.4	12.2 $\pm$ 0.8		11.5 $\pm$ 0.6	2.23 $\pm$ 1.26	154 $\pm$ 41	922 $\pm$ 227	1956 $\pm$ 168		
		35.2	11.2 $\pm$ 1.0		12.0 $\pm$ 0.6	1.47 $\pm$ 0.57	190 $\pm$ 46	681 $\pm$ 141	2006 $\pm$ 150		

<sup>1</sup> arrows indicate significant decrease in relation to the starting state; <sup>2</sup> different letters indicate significant differences at  $p < 0.05$ ; no letters—no significant differences for plant protection treatments or for P-fertilization treatments.

As in the case of Fe, P fertilization variants did not differentiate the content of total Al in the soil in the presented experiments, and the amounts of Al brought with P fertilizers (0.025–7.88 mg kg<sup>-1</sup> of soil, applied with PR at P dose of 17.6 kg ha<sup>-1</sup> and A<sub>s</sub>B<sub>m</sub> at the P dose of 35.2 kg ha<sup>-1</sup>, respectively; assumptions as specified in Section 3.1.) should be considered small against the initial Al content in the soil. However, it is thought that high concentrations of Al compounds may pose a greater threat to the environment than high concentrations of Fe compounds [83].

Similar to Fe, in the experiments established on acidic soils (experiments II–IV), a decrease in total Al content after harvesting the test plant was recorded in relation to the initial level, and this decrease was irrespective of P fertilization. It has long been known that the concentration of soluble Al increases rapidly in acidic soils (pH < 5.5) and that this soluble Al is potentially toxic for plant growth [39]. In the present study, the content of a mobile and bioavailable Al fractions in soil and fertilizers was not investigated. The amount of Al migrating deep into the soil profile [84] was also not studied.

Of the active forms of Al, some portion was taken up by field plants (wheat and weeds) and accumulated in their biomass, with the highest concentration of Al in post-harvest residues, followed by aboveground organs of weeds, less in wheat straw, and by far the least in wheat grain (Table 4). The Al content of these plant biomass components was not differentiated by P fertilization variants/treatments in any of the experiments conducted. In contrast, Kepka et al. [85] reported that the application of municipal sewage sludge to soil had a significant effect on increasing Al content in grain and straw of spring barley. In the present study, however, the main raw material of the biofertilizers tested was SSA, in which Al occurs typically in the form of anorthite and berlinite [86] and thus is hardly available to plants. Therefore, it can be inferred that the form and amount of Al entering the soil with the applied fertilizers did not contribute to the increase of bioavailable Al in the soil. Moreover, the activity of the P-solubilizing bacteria contained in the biofertilizers did not lower the soil pH, e.g., [87], which could promote the release of active Al into the soil solution and its uptake by plants.

In relation to the range given by Kabata-Pendias and Pendias [59] for cereal grains, as well as the results of other authors [85,88–90], the Al content in wheat grains in the present study should be assessed as particularly low. In Experiments II and III it was even below the level of detection. The values obtained in the present study correspond rather with those reported by Kolbaum et al. [91]. Although the beneficial effects of Al on plants are mentioned in the literature [39], even its indispensability [92], neither the deficiency level nor the range of its optimal content in plants has been established [59,85]. The issue of Al toxicity to plants and plant tolerance to Al has been addressed more frequently [39,93]. However, unlike many other PTEs (e.g., Cd, Pb), excessive or toxic Al content in plant biomass is not rigorously defined, since the extent of tolerance to excess Al and the ability to uptake and translocate it vary within plant species and even within cultivars of the same species [73]. In addition, despite its proven toxicity to animals and humans [94], current national and international legislation does not specify maximum levels of Al in cereals for food and feed purposes [60–64]. Earlier studies suggested 200 mg kg<sup>-1</sup> as the maximum allowable Al content for cereals [95]. Today, some authors also advise food safety authorities to set a maximum limit for the content of this element in cereals and cereal products [94].

The level of Al in aboveground weed biomass is not surprising when considering the range of values reported by Kabata-Pendias (X0–X00 mg/kg) [73], as well as the fact that some species of natural plants are known to accumulate more than 1000 mg kg<sup>-1</sup> dry mass of Al in their stems and leaves [96].

The high Al content in the biomass of post-harvest residues is a consequence of the Al concentration in plant roots. The root is seen to be the main region of Al accumulation in the plant, and the content of this metal in the root can be from several dozen up to 100 times higher than in shoots [77,90]. Root retention of Al, preventing its transport to the shoot, was proposed as one mechanism of plant tolerance to Al [97].

As in the case of Fe, in experiment II, higher Al content in weed biomass was found under PP+ conditions than under PP– conditions, which was also likely due to modification of the weed community structure under the influence of the applied herbicide. Surprisingly, there was no similar differentiation in Al content in post-harvest residues under the influence of the plant protection factor.

#### 4. Conclusions

The SSA-based biofertilizers containing P-solubilizing bacteria (*B. megaterium* or *A. ferrooxidans*) did not affect the total Fe and Al content in the soil or the concentration of these elements in plant biomass (wheat grain and straw, weeds, post-harvest residues) when applied at P doses up to 35.2 kg ha<sup>-1</sup>. The Fe and Al concentrations in the studied plant biomass components were arranged according to the following pattern: wheat grain < wheat straw < weed aboveground biomass < post-harvest residues. The tested preparations failed as a strategy to biofortify wheat grain in Fe, but Fe and Al levels in grain did not suggest a potential risk to consumers.

The results presented here, and the biofertilizers' yield-forming performance and other environmental effects, allow them to be seen as a promising approach to a sustainable circular P economy. However, further research on elemental mobility due to biological activators is advisable. Furthermore, long-term studies with repeated applications of SSA-based biofertilizers are still necessary.

**Supplementary Materials:** The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/agronomy12061475/s1>, Table S1: Elemental composition of SSA-based P-fertilizers; Table S2: Basic agricultural data for the experiments; Table S3: Soil characteristics before the start of the experiments; Table S4: Precipitation and air temperature during the study period according to the Meteorological Station in Bałcyny, Poland; Soil and plant sampling and preparation; Chemical analyses.

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