

Biosolids from Wastewater Treatment Plant as an Alternative for Soil Decontamination with Aldrin Agrochemical

Andrea del Pilar Gamarra Sánchez^a, Danny Lizarzaburu-Aguinaga^{b*}, Elmer Benites-Alfaro^a, Carlos Del Valle Jurado^c, Jorge Leonardo Jave Nakayo^c, Carlos Cabrera Carranza^c, Jorge López Bulnes^c, Guido Rene Suca-Apaza^d

^a Universidad César Vallejo, Av. Alfredo Mendiola 6232, Los Olivos, Lima, Perú

^b Universidad César Vallejo, Av. Argentina 1795, Callao, Perú

^c Universidad Nacional Mayor de San Marcos, Lima, Perú

^d Universidad César Vallejo, Carretera. Central Km 8.2 Ate, Perú

dlizarzaburu@ucv.edu.pe

Soil contamination by agrochemicals is a significant environmental problem that can negatively impact human health, biodiversity, and water quality. Excessive or inappropriate use of agrochemicals, including the pesticide Aldrin, commonly used as an insecticide in agricultural activity, can contaminate soil. The research aimed to use biosolids from a wastewater treatment plant to decontaminate agricultural soil containing Aldrin pesticide. For the investigation, soil samples were taken from the Carabayllo area, and 2 kg were placed in each container; then, biosolids from the wastewater treatment plant were added in proportions of 10, 20, and 30 %, respectively. The test was done in triplicate for each percentage. After 90 days of treatment, a 70 % decrease in the pesticide Aldrin was found, for a rate of 30 % of biosolid added to the soil. Therefore, it is established that sewage biosolids allow the reduction of the pesticide Aldrin, becoming an environmentally sustainable alternative to decontaminate soils.

1. Introduction

Aldrin (C₁₂H₈Cl₆) is a solid, non-flammable, insoluble, and highly persistent organochlorine chemical (Instituto Nacional de Seguridad e Higiene en el Trabajo, 2015). Due to its indiscriminate use, it has impacted agricultural soils near populated or industrialized areas of the capital of Peru. This pesticide was banned in 1991 by Supreme Decree No. 037-91-AG. However, it was used until 2004 (Consejo Nacional del Ambiente, 2006). Currently, direct effects are evident, such as the impact of heavy metals (Bonelli & Manni, 2019), reduction of agricultural capacity (Tesi et al., 2020), indirectly due to the application method, the wind rose, the temperature, the residues and their persistence (520 weeks) can generate health problems due to effects on microorganisms in the soil (Mehlhorn et al., 2023) and vectors (*Anopheles gambiae*), the impacts on the ecosystem may be related to bioaccumulation in fish (Unyimadu et al., 2018).

A possible solution to this problem is through various chemical methods, membrane separation, phytoremediation, bioremediation, incineration, ozonation, salt oxidation, Fenton oxidation, photocatalytic degradation, and nanotechnology (Kaur et al., 2022). Significant results have been evidenced with the use of a photocatalytic reactor for the reduction of atrazine and thiacloprid by more than 90% (Navarra et al., 2023), nanotechnology treatment for the removal of organochlorines (Sarno et al., 2017) using silver and graphene nanoparticles, a case of phosphorus nano treatment was also found using nano zero-valent iron (nZVI) achieving an absorption capacity of 68 mg/g (Bavasso et al., 2016), phytoremediation of *Ocimum basilicum* L and its rhizosphere for the organochlorine pesticide endosulfan, bioremediation with fungi to reduce organophosphates with results between 64% and 73% (Benites-Alfaro et al., 2023); With all this evidence, the idea of evaluating the application of biosolids from a wastewater treatment plant for the treatment of soil contaminated by Aldrin arose.

To do this, the physicochemical and microbiological parameters of the soil were determined before and after treatment to determine the reduction in pesticide concentration according to the best dose of biosolid, which allowed to demonstrate whether biosolids can improve the quality of agricultural soil in the Carabayllo area by improving its physicochemical properties and reducing existing chemical contaminants such as organochlorine compounds (Aldrin), reducing the need for fertilizer application (Ozores-Hampton & Mendez, 2010).

2. Methodology

The research was developed through a three-stage experimental process (Figure 1). In the first stage, soil preparation and sample extraction are carried out to determine the physicochemical and microbiological parameters and the concentration of organochlorine. The contaminated soil samples are mixed with the three treatments in their (three repetitions), remaining at rest for eight weeks. Finally, each treatment and repetition sample is collected and taken to the laboratory to validate the results.

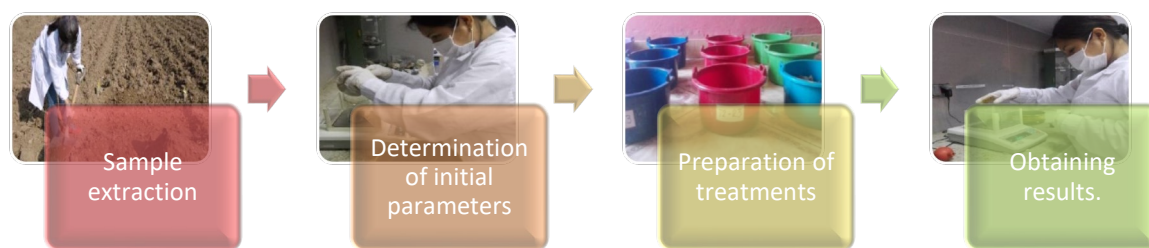


Figure 1: Experimental process

2.1 Extraction of soil samples contaminated with Aldrin

To collect the sample, the soil was extracted from an agricultural area located in the district of Carabayllo, province of Lima and department of the same name, at the coordinates E281092 - N8688543; the quartering method was used, stacking the sample until it formed a cake. The compact was divided into four equal parts, and the two ends were selected for the sample. This procedure was repeated until a weight of 20 kilograms was obtained for use in the experimental stage.

In addition, a 5 kg sample of sewage sludge dried over 22 months was taken near a wastewater treatment plant.

2.2 Determination of physicochemical and microbiological parameters.

Prior treatment was carried out on the sample in the laboratory; this consisted of grinding the sample until it was homogeneous, then the sample was dried in an oven at 60 °C for 30 minutes, and finally, the sample of contaminated soil was sieved in an ASTM No. 35 mesh, to remove all types of impurities. Subsequently, the physicochemical and microbiological characteristics and the concentration of Aldrin in the contaminated soil sample are determined. In the case of biosolid, the same treatment was also followed, except that the drying temperature was 90 °C, and it was sieved with an ASTM No. 135 mesh.

Subsequently, a 20 g sample of soil contaminated with Aldrin was taken to determine humidity following the ASTM D-2216-98 methodology, and the percentage of moisture was calculated with Eq(1).

$$\% \text{ moisture} = \frac{\text{wet weight} - \text{dry weight}}{\text{dry weight}} \times 100 \quad (1)$$

The soil pH analysis was done using a 50 g contaminated soil sample. Then, 50 ml of distilled water was added and homogenized for 30 seconds. It was allowed to stand for 3 minutes, and the process was repeated by preparing three samples. Ultimately, the pH was measured using a Hanna brand multiparameter instrument model HI98128. The same instrument also reads the electrical conductivity.

The determination of organic matter (using ASTM D-2974-00) began with sieving the contaminated soil after drying at 105 °C. The soil was then placed in an adequately tared crucible and weighed on the analytical balance (P1); the crucible with the sample was calcined at 430 °C for 48 hours. It was cooled in a desiccator for 20 minutes and weighed (P2). With Eq 2, the percentage of organic matter was calculated.

$$\% \text{ de M. O.} = \left[\frac{P1 - P2}{P1} \right] \times 100 \quad (2)$$

Soil texture was determined using the Bouyocus method (ASTM 152-H). A sample of 2 kg of soil was taken from the agricultural area of Carabayllo. The procedure was to use 50 g of soil mixed with 100 ml of water and 10 ml of the dispersing agent ((NaPO₃)₆), and it was taken to a mechanical agitator for 10 minutes; the suspension was transferred to a test tube (1000 ml), and it was gauged with distilled water, with a glass rod it was agitated again for 1 min. It was left to rest for 40 sec. Then the hydrometer of bouyocus is introduced to take the density reading (1st measurement); with a thermometer also, the temperature is measured to find the correction of the hydrometer of baoyocus with Eq (3). After 2 hours, the measurement is repeated (2nd measurement), and the correction is calculated. The calculation of the percentages of sand, silt, and clay is done with Eq (4), Eq (5), and Eq (6), whose values are shown in the results section and are taken to the texture diagram.

$$c = (T - 0.36) - 7 \quad (3)$$

c= will be the correction value added to the value measured by the hydrometer

T= temperature read

$$\% \text{ of sand} = 100 - \frac{1st \text{ corrected reading} * 100}{sample \text{ weight}} \quad (4)$$

$$\% \text{ of clay} = 100 - \frac{2nd \text{ corrected reading} * 100}{sample \text{ weight}} \quad (5)$$

$$\% \text{ of silt} = 100 - (\% \text{ of sand} + \% \text{ of clay}) \quad (6)$$

The characterization of actinomycetes was evaluated for microbiological evaluation because these filamentous bacteria (high positive) are widely distributed in the rhizosphere. Their properties have been studied for their ability to control pathogens focused on crops' bio-protection, which stimulates the reduction of pesticides (Franco-Correa, 2010).

For the analysis of the physicochemical characteristics of the biosolids, the same procedures were used, and as microbiological tests, fecal and total coliforms were determined to establish the suitability of the mixture with the contaminated soil and use as a treatment product, according to Supreme Decree No. 015- 2017-VIVIENDA (Ministerio de Vivienda, Construcción y Saneamiento, 2017). In addition, the microbial growth of actinomycetes was also evaluated for their ecological benefits as protectors of mycorrhiza; the test used was the colony-forming units (CFU) count.

2.3 Treatment process with biosolid.

For the experimental part, the treatments were defined by preparing containers containing doses with a percentage ratio of 10 % (222 g of biosolid), 20 % (500 g of biosolid), and 30 % (857 g of biosolid) that were mixed with samples of 2 kg of soil contaminated with Aldrin. Three repetitions for each percentage relationship were also defined (Figure 2), with nine samples that were then controlled and monitored.

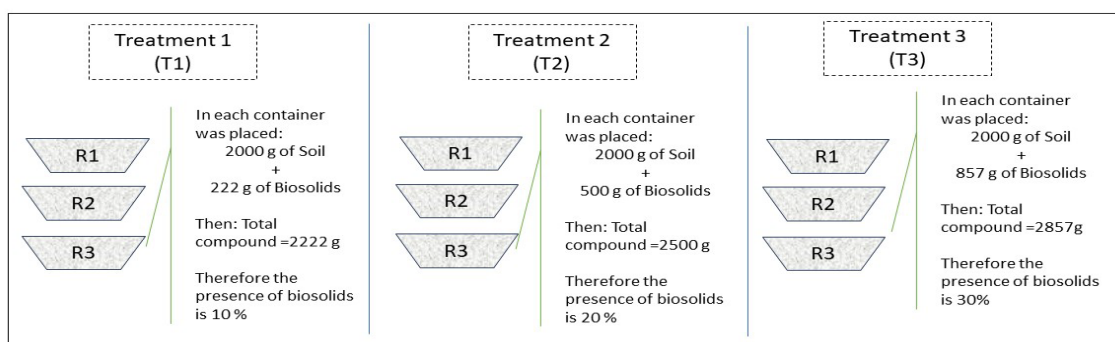


Figure 2: Biosolid treatments.

The treatments were coded as T1, T2, and T3 for the percentages of 10, 20, and 30 % of biosolids used in the containers with contaminated soil. The containers were placed in a dry place at room temperature for 8 weeks. The humidity and temperature parameters were monitored daily. The pH was evaluated twice a week, and the influence of the parameters on bioremediation was established—the value of organic matter to show an increase or reduction of microorganisms.

3. Result and discussion

3.1 Physicochemical analysis of contaminated soil before treatment.

The results of the agricultural soil contaminated with Aldrin were characterized in the laboratory of the Universidad César Vallejo, and the result is presented in Table 1. In the same way, the biosolid was also characterized for its physicochemical and microbiological properties of fecal coliforms and total coliforms to support its application on agricultural land according to national regulations (see Table 2).

Table 1: Initial physicochemical and microbiological characteristics of the soil.

Parameter	Unit	value
Conductivity electric	dS /m	1.65
pH	1-14	7.72
organic matter	%	1.84
moisture	%	13.64
	% Sand	36
Texture	% Silt	3. 4
	% Clay	30
Actinomycetes	UFC	1 x 10 ⁴

Table 2: Initial physicochemical and microbiological characteristics of the biosolid.

Parameter	Unit	Value
Conductivity electric	dS /m	21.60
pH	1-14	5.23
organic matter	%	11.79
moisture	%	7.27
Coliforms fecal	NMP/g	13x10 ¹²
Coliforms totals	NMP/g	15 x 10 ¹²

The initial results of contaminated soil parameters showed values of 1.65 dS/m for electrical conductivity considered as a very slightly saline soil, pH was classified as slightly alkaline, and moisture was low considering the soil texture which should be between 50 to 70% (Galindo et al., 2017), the texture turned out to be clay loam (USDA, 1999), the presence of organic matter allows evaluating the productivity and fertility of the soil and the value obtained was 1.84, it is considered as medium and beneficial for the soil (FAO, 2009). The Aldrin content was 3.01 mg/kg, as reported by an analysis performed in a specialized and certified laboratory (Environmental Testing Laboratory).

In the case of biosolids, the results showed that they were very saline, strongly acidic, with a very high level of organic matter; the microbiological analysis was less than 1×10^3 , and they were considered suitable for application in soils according to Peruvian regulations (<1000 NMP), so they should be subjected to a pre-treatment for their use (this was not done in this research because its objective was mainly focused on the reduction of the agrochemical Aldrin). The moisture percentage for both samples was low (ICA, 1992). This is justified because, as mentioned above, the biosolid was extracted from the water treatment plant and dried a long time before (22 months) for confinement (the samples for the investigation were taken from there).

3.2 Results of physicochemical and microbiological analyses after treatment with biosolids.

Table 3 shows the results of the parameters evaluated after treatment with biosolids (T1, T2, T3) with three replicates (R1, R2, R3); the averages for each parameter and the absolute error of the measurements are calculated.

Table 3: Physicochemical and microbial characteristics after treatment.

Treatment	Parameter	R1	R2	R3	Average	Absolute Error
T1	pH	8.03	7.15	7.5	7.56	0.31
	organic matter (%)	13.2	14.1	15.11	14.14	0.65
	moisture (%)	8.13	8.78	10.12	9.01	0.74
T2	pH	7.88	8.49	8.18	8.18	0.20
	organic matter (%)	18.08	17.55	17.64	17.76	0.22
	moisture (%)	14.65	15.77	15.96	15.46	0.54
T3	pH	9.14	8.77	8.5	8.80	0.22
	organic matter (%)	13.93	18.25	17.32	16.50	1.71
	moisture (%)	14.68	18.39	20.25	17.77	2.06

Table 3 shows a progressive increase in pH from the treatment with 10, 20 and 30 % of biosolids used in the treatment; the third treatment, whose average value is 8.80, is the most basic; this value is within the range suitable for soil bioremediation (Tibamba et al., 2024); Likewise, the results of organic matter of the soil treated with biosolids show a significant increase for the second treatment, this shows that the application of biosolids improves soil conditions which allows it to be used as fertilizer, 2024); Likewise, the results of organic matter of

the soil treated with biosolid show a significant increase for the second treatment, this shows that the application of the biosolid improves soil conditions which allows it to be used as fertilizer or soil amendment material (Pulgarín Muñoz et al., 2022); the percentage of humidity also showed a significant increase due to the addition of the biosolid (Mansur Aisse, 2020). Microbial growth was also evident, with growth being 3.2×10^4 , 3.6×10^4 and 4.0×10^4 CFU for treatments T1, T2, and T3, respectively. These results allow us to infer that this helped protect the root and improve the availability of nutrients in the soil, generating a direct positive impact in its application of agricultural activity (González Jiménez, 2010).

3.3 Reduction of Aldrin with biosolids treatment.

The effect of applying the biosolid at different doses to reduce the concentration of Aldrin in the contaminated soil was evaluated at doses of 10%, 20%, and 30%, with three repetitions, which allowed verifying the research objective (see Table 4). It should be noted that before treatment, the concentration of Aldrin in the contaminated soil was evaluated at 3.01 mg/kg and that no blank test was performed to evaluate the degradation of the contaminant without the additives.

Table 4: Aldrin reduction according to treatment

Treatment	Aldrin Concentration	R1	R2	R3	Average	Absolut mistake	Reduction (%)
T1	Aldrin (mg/kg)	2.4	2.4	2.39	2.40	0.00	20
T2	Aldrin (mg/kg)	1.32	1.34	1.31	1.32	0.01	56
T3	Aldrin (mg/kg)	0.93	0.9	0.91	0.91	0.01	70

Initially, the Aldrin concentration was 3.01 mg/kg in dry mass. The concentration of the contaminant in the samples treated with biosolids decreased according to the percentage of biosolids applied, demonstrating the effectiveness of biosolids as a carbonaceous agent in improving contaminated soil (Saito et al., 2011), complying with the soil quality standard of the Peruvian regulations (Ministry of Environment, 2013). The pollutant Aldrin was reduced in all replicates, with the highest concentration reduction in treatment T3. The mechanism for the reduction of Aldrin was due to the adsorption capacity of biosolids, facilitated by the presence of organic matter, which absorbs organochlorine compounds such as Aldrin, which have low solubility in water but high disposability (so they are prone to accumulate in animal and plant fatty deposits); This adsorption can be influenced by factors such as pH, particle size, temperature and contact time (Espinoza et al., 1995).

The study demonstrates that biosolids can improve the quality of soil contaminated with Aldrin by reducing its concentration. Therefore, tests are continuing to determine the saturation point of the biosolids at the wastewater treatment plant in the Carabayllo area, which is the optimal percentage for its use instead of being disposed of as waste.

4. Conclusion

Based on the results obtained, it was established that the behavior of the biosolids with each treatment dose (10 %, 20 %, and 30 %) reduces the Aldrin contaminant in the agricultural soil, and the effect of this element on the microorganisms present in the soil (actinomycetes) increases the capacity of this bioremediation to improve the physicochemical properties of the soil; however, it is the treatment with 30 % of biosolids that obtained the best results, reducing 70 % of the agrochemical Aldrin. This result will allow future experimental field-level trials to remediate and improve the quality of agricultural soils degraded by these fertilizers.

Acknowledgments

The authors thank the Vice-Rectorate for Research of César Vallejo University's financial support in disseminating this scientific work and the Research Group on Environmental Management and Solid Waste (GIGATRE) for supporting the article.

References

- Arias J. A., Rojas D., Dierkmeier G., Riera C., & Cabrera N., 1990. Surveillance Series 9: Organochlorine Pesticides, [in Spanish], Pan American Center for Human Ecology and Health.
- Bavasso I., Vilardi G., Stoller M., Chianese A., & Di Palma L., 2016. Perspectives in nanotechnology-based innovative applications for the environment. *Chemical Engineering Transactions*, 47, 55–60. <doi.org/10.3303/CET164700> accessed 05.01.2024.

- Benites-Alfaro E., Silva L. M., Guanilo A. P., Nakayo J. J., Castañeda-Olivera, C. A., Lizarzaburu-Aguinaga D. A., & Cabrera C. F., 2023, Pleurotus Ostreatus and Trametes Versicolor Fungi to Decontaminate Soils Containing Organophosphates Methamidophos and Cadmium. *Chemical Engineering Transactions*, 100(March), 97–102. <doi.org/10.3303/CET23100017> accessed 14.01.2024.
- Bonelli M. G., & Manni A., 2019. Organochlorine pesticides (OCPs) forecasting from heavy metals determinations. *The Seventh International Conference on Environmental Management, Engineering, Planning & Economics*, May.
- Consejo Nacional del Ambiente (CONAM)., 2006, *Inventario Nacional de Plaguicidas COP*.
- United States Department of Agriculture (USDA)., 1999, Guide for the Evaluation of Soil Quality and Health .
- Derco J., Dudáš J., Šilhárová K., Valičková M., Melicher M., & Luptáková A, 2012. Removal of selected micropollutants by ozonation. *Chemical Engineering Transactions*, 29 (2012), 1315–1320.
- Díaz J and Cifuentes G., 2020, From generation to sustainable use: sludge and biosolids from water and wastewater treatment, [in Spanish], Saneamiento de Lodos y Biosólidos (Issue December), Universidad de Boyacá. <doi.org/10.24267/9789585120136> accessed 17.01.2024.
- Espinosa L., Ramírez G. and Campos N., 1995, Analysis of organochlorine residues in the sediments of mangrove areas in the Ciénaga Grande of Santa Marta And Chengue Bay, Colombian Caribbean, [in Spanish], *Boletín de Investigaciones Marinas y Costeras - INVEMAR*, 24(1), 79-94.
- Franco-correa M., 2010, Use of actinomycetes in processes biofertilization. *Rev.Peru.Biol.*, 16(2), 239–242.
- González Jiménez Y. T., 2010, Actinomycetes: A View as Plant Growth Promoters [in Spanish], In Pontifical Javeriana University.
- Instituto Colombiano Agropecuario (ICA)., 1992, Fertilization in various crops; fifth approximation [in Spanish], (pp. 1–72). ICA.
- Instituto Nacional de Seguridad e Higiene en el Trabajo, 2015, DLEP 95 Aldrin. <www.insst.es/documents/94886/288875/DLEP+95+Aldrin.pdf/97b85590-237c-40b8-9a15-232f0eb1a968> accessed 12.01.2024.
- Kaur K., Rana A. K., Kumar B., Kumar V., & Saruchi., 2022, Advance remediation technologies for the removal of organochlorine from water and wastewater. In *Pesticides Remediation Technologies from Water and Wastewater*. INC. <doi.org/10.1016/B978-0-323-90893-1.00014>.
- Ministerio de Vivienda Construcción y Saneamiento., 2017, Supreme Decree that approves the Regulation for the Reuse of Sludge generated in Wastewater Treatment Plants, [in Spanish], *El Peruano*, (pp. 32–40).
- Ministerio del Ambiente., 2013, Environmental Quality Standards (EQS) for Soil, [In Spanish], In *El Peruano* (pp. 491497–491500).
- Navarra W., Sacco O., Vaiano V., & Venditto V., 2023, Pesticides Removal from Wastewater using a Pilot-scale Photocatalytic Reactor. *Chemical Engineering Transactions*, 98(December 2022), 159–164.
- Ozores-Hampton M., and Mendez J., 2021, Use of Biosolids in Vegetable Production [in Spanish], University of Florida/IFAS, <edis.ifas.ufl.edu/publication/HS1183>.
- Organización de las Naciones Unidas para la Alimentación y la Agricultura (FAO)., 2009, Guide to soil description [in Spanish].
- Pulgarin C. E., Saldarriaga J. C., & Correa M. A., 2022, Analysis and perspectives of anaerobic treatment and use of biological sludge in Latin America, [In Spanish], *Jornal EIA*, 19(38), 1–6. <doi.org/10.24050/reia.v19i38.1516> accessed 12.01.2024
- Ramirez-Sandoval M., Muñoz-Hernández S., & Velázquez-Fernández J. B., 2013, Mechanisms of phytoremediatory effect of ocimum basilicum l. and its rhizosphere exposed to different concentrations of the organochlorine pesticide endosulfan. *Chemical Engineering Transactions*, 34(April), 73–78. <doi.org/10.3303/CET1334013> accessed 15.01.2024.
- Saito T., Otani T., Seike N., Murano H. & Okazaki, M., 2011. Suppressive effect of soil application of carbonaceous adsorbents on dieldrin uptake by cucumber fruits. *Soil Science and Plant Nutrition*, 57(1), 157–166. <doi.org/10.1080/00380768.2010.551281> 15.01.2024
- Sarno M., Casa M., Cirillo C., & Ciambelli P., 2017, Complete removal of persistent pesticide using reduced graphene oxide-silver nanocomposite. *Chemical Engineering Transactions*, 60, 151–156. <doi.org/10.3303/CET1760026> accessed 15.01.2024.
- Tesi J. N., Tesi G. O., Ossai J. C., & Agbozu I. E., 2020, Organochlorine pesticides (OCPs) in agricultural soils of Southern Nigeria: spatial distribution, source identification, ecotoxicological and human health risks assessment. *Environmental Forensics*, 0(0), 1–13. <doi.org/10.1080/15275922.2020.1850570> accessed 14.01.2024.
- Tibamba T., Wang Y., Gameli R. B. H., Mbage B., Duwiejuah A. B., Wang N., & Bing L., 2024, Mono and Simultaneous Adsorption of Aldrin and Toxic Metals from Aqueous Solution Using Rice Husk-Biochar. *BioResources*. <doi.org/10.15376/biores.19.1.257-275> accessed 12.01.2024