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

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Soil contamination by agrochemicals is a significant environmental problem that can have negative impacts on 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 result in soil contamination. The objective of the research was to use biosolids from a wastewater treatment plant to decontaminate agricultural soil containing the pesticide Aldrin. 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 percentage 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

Agricultural soils near the capital of Peru usually contain different contaminants due to the indiscriminate use of pesticides or their proximity to populated or industrialized areas. One of these contaminants is Aldrin (C12H8Cl6), a solid, non-flammable organochlorine, insoluble in water and highly persistent (Instituto Nacional de Seguridad e Higiene en el Trabajo National, 2015); which has been used as a pesticide until 1991 when, by Supreme Decree No. 037-91-AG, its import and registration was prohibited, however its use and application has continued illegally until the end of 2004 (Consejo Nacional del Ambiente, 2006); showing that the impact on soils due to heavy metals (Bonelli & Manni, 2019) and their agricultural capacity persist to this day.

Aldrin contamination in soils can occur directly due to agricultural and health use (Tesi et al., 2020) and indirectly caused by the method of application, the wind rose, high temperatures, solid waste, among others, all this combined with its high persistence (520 weeks of activity), its bioaccumulation factor (4,444 in fish), the resistance that vectors can generate (Anopheles gambiae) by adaptive phenomena or effects on microorganisms (mites, nematodes, worms, insect larvae and bacteria) in the soil (Arias et al., 1990).

Faced with such a problem, it is possible to address the solution through various chemical methods, membrane separation, phytoremediation, bioremediation, incineration, ozonation, oxidation with salts, oxidation with Fenton, photocatalytic degradation, nanotechnology (Kaur et al., 2022), Within the collection, important results have been found such as the use of a photo-catalytic reactor for the reduction of atrazine and thiacloprid by more than 90% (Navarra et al., 2023), treatment with nanotechnology for the removal of organochlorines (Sarno et al., 2017) using silver and graphene nanoparticles, a case of phosphorus nano treatment using zero-valent nano 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, finding that more research is still required (Ramirez-Sandoval et al., 2013), the use of ozone in the case of wastewater, eliminating between 70 and 90% of the organochlorines (Derco et al., 2012); bioremediation with fungi to reduce organophosphates with results between 64 % and 73 % (Benites-Alfaro et al., 2023); Taking into consideration all these results, the need arises to evaluate the application of biosolid from a wastewater treatment plant for the treatment of soil contaminated by Aldrin. For this, the physicochemical and microbiological parameters of the soil before and after treatment were determined and the optimal dose of the biosolid to reduce the concentration of the pesticide.

The research was motivated in order to determine whether biosolids that are exposed as waste in the vicinity of a wastewater treatment plant have the ability to improve the environmental quality of agricultural soil in the area of Carabayllo, since by scientific theory biosolids improve soil properties, reduce the application of fertilizers by providing physical and chemical benefits and especially reduces existing chemical pollutants such as organochlorine compounds such as Aldrin (Ozores-Hampton M., and Mendez J., 2021).

* 1. Methodology

The research was developed through a three-stage experimental process (figure 1), in the first the sample extraction is carried out, to later determine the physicochemical and microbiological parameters and the concentration of the organochlorine, followed by the preparation of the soil. contaminated with the treatments (3 treatments and 3 repetitions) remaining at rest for eight weeks, and finally, samples of each treatment are collected to take them to the laboratory and obtain the results.

Figure 1: Experimental process

* + 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. compact that was divided into four equal parts, the two ends were selected for the sample, this procedure was repeated until a weight of 20 kilograms was obtained to be used in the experimental stage.

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

* + 1. 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 ºC for a period of 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 ºC 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, the percentage of moisture was calculated with Eq(1).

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| $$\% moisture= \frac{wet weight-dry weight}{dry weight}x100$$ | (1) |

The soil pH analysis was carried out by taking a 50 g sample of contaminated soil, then 50 ml of distilled water was added and homogenized for 30 seconds, allowed to stand for 3 minutes and the process was repeated by preparing 3 samples. At the end the pH was measured using a Hanna brand multiparameter instrument model HI98128. With the same instrument, the electrical conductivity was read.

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

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| --- | --- |
| $$\% de M.O. = \left[\frac{P1-P2}{P1}\right] x 100$$ | (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 a period of 1 min and it was left to rest for 40 sec. Then the hydrometer of bouyocus is introduced to take the reading of the density (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 also 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.

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| $$c=\left(T-0.36\right)-7$$c= will be the correction value added to the value measured by the hydrometer T= temperature read | (3) |
| $$\% of sand=100-\frac{1st corrected reading\*100}{sample weight}$$ | (4) |
| $$\% of clay=100-\frac{2nd corrected reading \*100}{sample weight}$$ | (5) |
| $$\% of silt=100-(\% of sand+\% of clay)$$ | (6) |

For the microbiological evaluation, the characterization of actinomycetes was evaluated, because these filamentous bacteria (high positive) are widely distributed in the rhizosphere, whose properties have been studied for their ability to control pathogens focused on the bio-protection of crops, that allows stimulating 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, Costrucción 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.

* + 1. 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. It was also defined that there were three repetitions for each percentage relationship (Figure 2), with a total of 9 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 the bioremediation was established. the value of organic matter to show an increase or reduction of microorganisms.

* 1. Result and discussion
		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 with the result presented in Table 1. In the same way, the biosolid was also characterized for its physicochemical and microbiological properties of both 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.

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| --- | --- | --- |
| **Parameter** | **Unit** | **value** |
| Conductivity electric | dS /m | 1.65 |
| pH | 1-14 | 7.72 |
| organic matter | % | 1.84 |
| moisture | % | 13.64 |
| Texture | % Sand | 36 |
| % Silt | 3. 4 |
| % Clay | 30 |
| Actinomycetes | UFC | 1 x 104 |

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 | 13x1012 |
| Coliforms totals | NMP/g | 15 x 1012 |

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, 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 reported by 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 1x103 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).

* + 1. Results of physicochemical and microbiological analyses after treatment with biosolids.

The results of the parameters evaluated after treatment with biosolids (T1, T2, T3) with 3 replicates (R1, R2, R3) are shown in Table 3, their 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 | Absolut mistake |
| 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 bio-remediation (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 shows 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 (Pulgarin 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 x 104, 3.6 x 104 and 4.0 x 104 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).

* + 1. Reduction of Aldrin with biosolids treatment.

The effect of applying the biosolid at different doses for the reduction of Aldrin in the contaminated soil was evaluated, which were doses of 10 %, 20 %, and 30 %, with three repetitions, which allowed the objective of the research to be verified (see Table 4), it is necessary to indicate that before treatment, the concentration of Aldrin in the contaminated soil was evaluated as 3.01 mg/kg.

Table 4: Reducción de Aldrin según tratamiento

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Treatment | Aldrin Concentration | R1 | R2 | R3 | Average | Absolut mistake | Reduction (%) |
| T1 | Aldrín (mg/kg) | 2.4 | 2.4 | 2.39 | 2.40 | 0.00 | 20 |
| T2 | Aldrín (mg/kg) | 1.32 | 1.34 | 1.31 | 1.32 | 0.01 | 56 |
| T3 | Aldrín (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 and 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 to improve contaminated soil (Saito et al., 2011), complying with the soil quality standard of the Peruvian regulations (Ministry of Environment, 2013). In all replicates there was a reduction of the pollutant Aldrin, 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 liposubility (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 the biosolid allows for improving the quality of soil contaminated with Aldrin, reducing its concentration; Therefore, tests are continuing to determine the saturation point of the biosolid of the wastewater treatment plant in the Carabayllo area, that is, the optimal percentage for its use, instead of being disposed of as waste.

* 1. 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 bioremediator 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 trials at the experimental field level to remediate and improve the quality of agricultural soils degraded by these fertilizers.

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References

Arias Verdes J. A., Rojas Companioni D., Dierkmeier Corcuela G., Riera Betancourt C., & Cabrera Cruz 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/CET1647010˃ accessed 05.01.2024.

Benites-Alfaro E., Silva Carhuatocto L. M., Guanilo Iñigo A. P., Nakayo J. J., Castañeda-Olivera, C. A., Lizarzaburu-Aguinaga D. A., & Cabrera Carranza 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*.

Departamento de Agricultura de Estados Unidos de Norteametica (USDA)., 1999, Guide for the Evaluation of Soil Quality and Health [in Spanish].

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 Bocayá. <doi.org/10.24267/9789585120136˃

Espinosa, L., Ramírez G. and Campos N., 1995, Analysis of organochlorine residues in the sediments of mangrove areas in the Cénaga 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 Costrucción 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 Muñoz C. E., Saldarriaga Molina J. C., & Correa Ochoa 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˃

Ramirez-Sandoval M., Muñiz-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˃

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 Matthew 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˃