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Research Article

# Effect of exogenous organic carbon on sorption of atrazine herbicide in two contrasting soils of Rawalakot and Attock

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## Abstract
Pesticides are chemical substances employed in agriculture to manage crop-damaging pests like insects, weeds, and diseases. The global use of pesticides has risen in response to the growing human population. Farmers aim to maximize their agricultural output by using significant quantities of pesticides. Nonetheless, this practice raises environmental concerns due to the persistent nature of pesticides and their interactions with diverse soil conditions. The presence of pesticides in soils is influenced by factors such as soil organic matter (SOM), clay content types, their quantity, and soil pH. The objective is to analyze how atrazine herbicide interacts with agricultural soils in Azad Jammu & Kashmir and Attock, which exhibit significant differences. Additionally, the study aims to explore the impact of added organic carbon on the sorption characteristics of atrazine herbicide in two contrasting soils selected from Azad Jammu & Kashmir (Rawalakot) and Attock (Punjab). These soils were amended with different types of organic sources such as biochar, compost, poultry manure, and FYM and sewage sludge. The sorption coefficient, $K_{d,}$ values were found to be affected by soil organic carbon and the accumulation of different sources to soil also influenced the sorption. AJK soil has organic carbon 7.6 g kg-$^{1}$ whereas Attock soil has 1.7 g kg-$^{1}$ and $k_d$ value for these soil has 11.69 Lkg$^{-1}$ and 4.67 Lkg$^{-1}$, respectively. Higher value of AJK un-amended was due to the presence of high organic carbon 7.6 g kg-$^{1}$ and low soil pH as compared to Attock un-amended 1.7 g kg-$^{1,}$ respectively. Overall positive relation was observed in all treatments with the addition of organic sources. Maximum value of $K_d$ observed were 16.11 LKg$^{-1}$ in AJK soil and it was 8.49 LKg$^{-1}$ in Attock soils. The sorption normalized on the basis of per unit organic carbon ($K_{oc}$) was 2120 LKg$^{-1}$ in AJK soil and in Attock soil 4999 LKg$^{-1}$, respectively. The current study showed that soils of AJK had more sorption capacity of atrazine as compared to Attock soil. The difference in their sorption capacity of atrazine in the soils (AJK & Attock) was due to differences in their mineralogy and soil chemicals properties including soil pH, SOM which are considered the key factors for the atrazine sorption. Therefore, it was concluded from these findings that rate and amount of pesticides applied to soils must be regarded soil chemicals and physicals properties which effect its movement into soils. The study also suggests that in order to reduce the soil amendments with the organic sources play as key role.

**Keywords**: Atrazine sorption; Chemicals; Herbicides; Pesticides.

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## Introduction

Pesticides are the most important agrochemicals used to control agricultural losses triggered by weeds, insects, and pests. The wide usage of pesticides in agriculture greatly influence in production of good quality food. However in addition to this haphazard usage of these chemicals have serious threats to the environment as they cause pollution due to their persistence. Pesticide residues have the potential to pose future threats to human health, as they can accumulate in the food chain and lead to extensive environmental contamination. The acute and chronic impacts of pesticide contamination can result in a range of adverse effects on human health, including carcinogenic, oncogenic, genotoxic, and teratogenic consequences (Singh *et al.*, 2018; Song *et al.*, 2012). Currently, in modern agriculture, the use of pesticides is considered an important part of agriculture system. Production of quality crops and food products can be increased with the use of pesticides in agricultural crops (Ahmad *et al.*, 2001). According to an estimate, during the 1960s the pesticide use in agriculture was 7 million tons per year, which jumped to 16226 million tons in 1977 (Baloch, 1985). Globally 3 million metric tons of pesticides are utilized every year that include 45% in Europe, 25 % in the USA, and 25 % in the remaining countries of the world (Popp, 2011).

Pakistan embarked upon using pesticides in 1964 with a meager use of 254 metric tons of formulated pesticides (Tariq *et al.*, 2007) which has currently reached to 90676 metric tons. Global use of these chemicals, herbicides share about 47.5 %, insecticides contribute to 29.5 %, and 17.5 % is the contribution of fungicides, whereas rest of the chemicals account for 5.5 % (Sharma *et al.*, 2020). Without use of these pesticides it is difficult to attain maximum production of agricultural crops (Pathak *et al.*, 2022). It is helpful to bear high and stable crops yield (Gianessi and Reigner, 2007) and decrease world starvation (Borlaug, 1968).

Among all herbicides triazine family has been more widely utilised in last ten years (LeBaron *et al.*, 2008). Worldwide, in more than 100 countries many triazine herbicides are currently recorded to provide weed control in a variety of crops. It has been mostly used in agricultural crops to minimize the effect of unwanted plant in various crops, such as maize, sugarcane and cereal crops. Due to its distinctive chemical characteristics of long time persistence in fields, soils and fresh water reported it is of major concern for environment sustainability (Clark and Goolsby, 2000; Blanchoud *et al.*, 2007). Among triazine family, atrazine (6-choloro-$N_2$ ethyl $N_4$-isopropyl 1, 3, 5-triazine-2, 4-diamine) is herbicide widely used in agriculture for pre-emergence regulation of broadleaf weeds and annual grasses. It has been the most widely used herbicide (about 30,000 tons annually) for maize crop (Hase *et al.*, 2008).

Sorption is the major processes which decreases pesticides movement in agricultural soils. Key factors regulating sorption of herbicides include hydrophobicity of the herbicide, the soil organic carbon (SOC) content, pH, type and amount of clay. Sorption is characterized by partition constant, $K_d$ (Wauchope *et al.*, 2002). Organic modifications, utilized to improve the organic carbon content of soils. Soil surface and subsurface materials can be altered that increases sorption potential and reduces pesticide contamination of ground water (Zsolnay, 1992). Exogenous organic matter addition to soil possibly a useful method to affect the sorption that may subsequently have impact on other processes such as degradation and leaching (Sharma *et al.*, 2016). To improve soil properties for increasing sorption of organic chemicals, soil amendments are necessary. However, only a little information is available on impact of exogenous organic carbon on sorption of atrazine herbicide and best of knowledge no such report is



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available for Pakistani soils. Hence, the current study was conducted with the objectives of assessing the sorption characteristics of atrazine herbicide in two dissimilar agricultural soils from Azad Jammu & Kashmir and Attock, as well as exploring how the introduction of external organic carbon impacts atrazine's sorption tendencies.

## Methodology

Two different types of agricultural soils sampled from Azad Jammu & Kashmir (Rawalakot) and Attock (Punjab) were air dried, sieved and added with different types of organic materials. Sewage sludge was collected from Dhok Kala Khan, Poultry Manure was collected from Poultry Research Institute, Rawalpindi whereas compost, biochar and farm yard manure were collected from PMAS-Arid Agriculture University Rawalpindi for the current study comprises of six treatments with three replications.

**Sample Preparation and soil analysis**

The soil samples were amended with biochar, poultry manure, sewage sludge, farm yard manure, and compost at the rate of 20 ton $ha^{-1}$. Soils were analyzed for the following parameters before treatments to measure soil physical and chemicals properties comprising soil texture, pH, electrical conductivity (EC), bulk density (BD), cation exchange capacity (CEC), and total organic carbon (TOC %).

**Atrazine sorption equilibria**

To establish the equilibrium duration for atrazine within the soil system, 2 grams of air-dried soil were placed in Kimax glass containers. Subsequently, 10 mL of a 1.5 mg L-1 atrazine solution in 0.01 M CaCl2 was introduced. Duplicate samples, along with a blank control, were vigorously agitated for durations of 0.5, 1, 2, 4, 6, 8, 12, 16, 20, and 24 hours. Following this, the containers were subjected to centrifugation at 2000 rpm for 20 minutes, and approximately 1 mL of the resulting supernatant was filtered using 0.45 μm Minisart filters. Atrazine concentrations were subsequently analyzed using High-Performance Liquid Chromatography (HPLC) equipped with UV/Vis detection at a wavelength of 220 nm. This analytical procedure allowed for the determination of the equilibrium time.

**Atrazine Sorption**

Atrazine sorption in soils were determined at temperature 24 ± 2 $^{o}$C following batch equilibrium method (Ahmad *et al.*, 2001). Sorption was determined by using 0.01 M $CaCl_2$.

**Atrazine determination**

Determination of atrazine in two amended and un-amended soils were made. Methanol and HPLC grade water (60:40 methanol: water) were used to prepare mobile phase. In the graduated cylinder the 600 mL of methanol and 400 mL of HPLC grade water was added and solution was made in the flask and was used for the determination of atrazine.

Stock solution of 100 mg $L^{-1}$ was made by taking 10 mg standard atrazine chemical in 100 mL volumetric flask and brought to volume with methanol. The atrazine standard solutions were prepared as under: 0.5 mg $L^{-1}$ standard solution: Add 0.5 mL of standard stock solution in to 100 mL of volumetric flask. Volume was made with mobile phase (60:40 methanol: water) to 100 mL. 1.0 mg $L^{-1}$ standard solution: Add 1 mL of standard stock solution in 100 mL of volumetric flask. Volume was made with mobile phase (60:40 methanol: water) to 100 mL. 1.5 mg $L^{-1}$ standard solution: Add 1.5 mL of standard stock solution in 100 mL of volumetric flask. Volume was made with mobile phase (60:40 methanol: water) to 100 mL. 2.0 mg $L^{-1}$ standard solution: Add 2.0 mL of standard stock solution in 100 mL of volumetric flask. Volume was made with mobile phase (60:40



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methanol: water) to 100 mL. 3.0 mg $L^{-1}$ standard solution: Add 3.0 mL of standard stock solution in 100 mL of volumetric flask. Volume was made with mobile phase (60:40 methanol: water) to 100 mL.

For different concentrations of atrazine standard curve was made. Sorption coefficients were determined by single solution concentration of each herbicide. The unit of $K_d$ value is L $kg^{-1}$. $K_d$= amount sorbed on soil (mg/kg) /amount in solution (mg/L)

**Statistical analysis**

The data that were collected underwent analysis of variance (ANOVA) using the statistical software "Statistix 8.1 version," as described in the study by Reddy et al. in 2019. To assess the significance of the treatment means, Tukey's Honest Significant Difference test was employed at a significance level of 5%.

## Results

**Sorption coefficient ($K_d$ and $K_{oc}$) of atrazine in soils**

Sorption coefficients ($K_d$ and $K_{oc}$) of two soils, Azad Jammu & Kashmir (Rawalakot) and Attock (Punjab), are shown in Figure 1. High value of $K_d$ (11.69 L $kg^{-1}$) was found in AJK (Rawalakot) un-amended soil as compared to Attock (Punjab) un-amended soil (4.67 $Lkg^{-1}$) whereas the $K_{oc}$ values of these soils were as 1338 $Lkg^{-1}$ and 2748 $Lkg^{-1}$, respectively (Figure 1). The obtained values of Sorption coefficients ($K_d$ and $K_{oc}$) were in line with the reference values in different soils already reported in the literature (Table 1).

Table 1. $K_d$ values for atrazine reported in different soils in literature.

| Sorption Coefficient ($K_d$) L $Kg^{-1}$ | References |
|---|---|
| 3.2-8.4 | Novak *et al.* (1997) |
| 1.7-4.7 | Weber *et al.* (2003) |
| 1.0-5.5 | Socías-Viciana *et al.* (1999) |
| 0.5 -20 | Schwab *et al.* (2006) |
| 0.7-52.1 | Ahmad *et al.* (2023) |
| 4.67-11.69 (un-amended) | Present study |

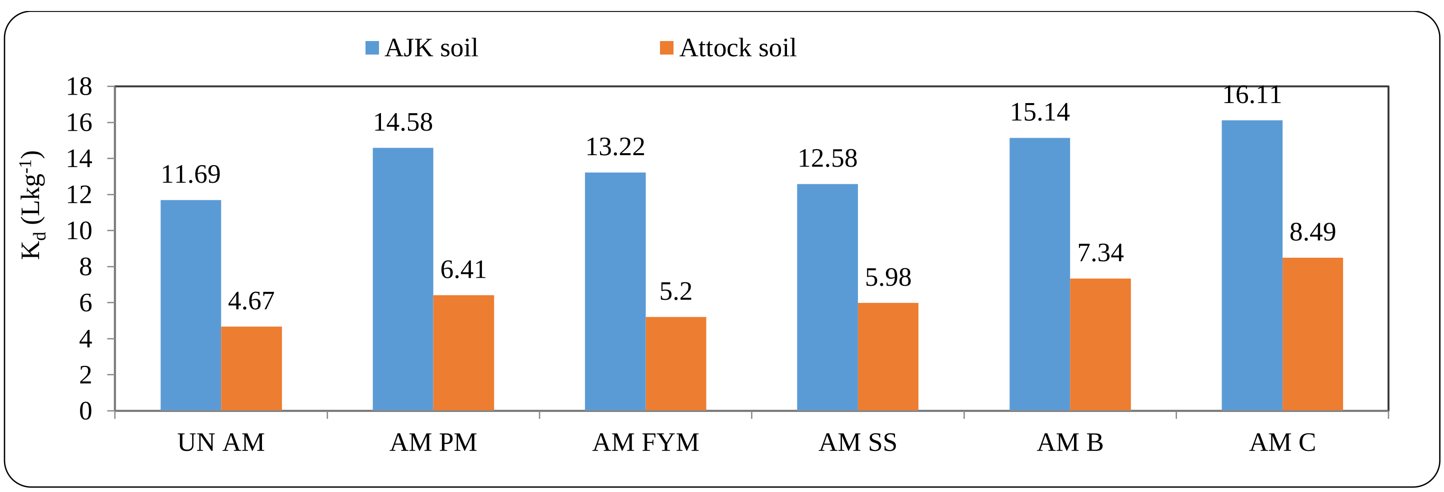


Figure 1. Sorption coefficient, $K_d$, of Atrazine for two amended and un-amended soils.
UN AM = UN-Amended, AM PM = Amended Poultry Manure, AM FYM= Amended Farmyard Manure, AM SS= Amended sewage sludge, AM B= Amended biochar, AM C= Amended Compost


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**Effect of Organic Carbon on Sorption of Atrazine**

SOC contents are considered the most significant features for the sorption of atrazine in soil which indirectly relates to the amount of organic matter.

The sorption coefficient, $K_d$, values were found to be affected by soil organic carbon and the accumulation of different sources to soil also influenced the sorption. AJK soil has organic carbon 7.6 g kg$^{-1}$ whereas Attock soil has 1.7 g kg$^{-1}$ and $k_d$ value for these soil has 11.69 LKg$^{-1}$ and 4.67 LKg$^{-1}$, respectively. Higher value of AJK un-amended was due to the presence of high organic carbon 7.6 g kg$^{-1}$ and low soil pH as compared to Attock un-amended 1.7 g kg$^{-1}$, respectively.

Overall positive relation was observed in all treatments with the addition of organic sources. The difference in $K_d$ values was due to the nature and types of organic carbon present in different sources. The results showed that highest impact on the sorption was observed in the soils whereas compost was used it may be due to the more stable and fine size particles. Maximum value of $K_d$ observed were 16.11 LKg$^{-1}$ in AJK soil and it was 8.49 LKg$^{-1}$ in Attock soils. The sorption normalized on the basis of per unit organic carbon ($K_{oc}$) was 2120 LKg$^{-1}$ in AJK soil and in Attock soil 4999 LKg$^{-1}$, respectively. It was observed that soil organic carbon had great influence on the sorption of atrazine in both soils, soil pH and clay content also influenced the sorption of atrazine.

The $K_d$ values of atrazine for un-modifed and modified soils with biochar shown in Table 4 and 5. The value of $K_d$ for AJK un-amended soil was 11.69 LKg$^{-1}$ whereas Attock soil had $K_d$ value of 4.67 LKg$^{-1}$. After biochar amendment in both soils $K_d$ values jumped to 15.14 LKg$^{-1}$ and 7.34 LKg$^{-1}$ for the respective soils.

Table 2. Atrazine Sorption Coefficient ($K_d$ & $K_{oc}$) for AJK Soil.

| Soils | $K_d$ (L kg$^{-1}$) | $K_{oc}$ (L kg$^{-1}$) |
|---|---|---|
| AJK (UN AM) | 11.69 | 1338 |
| AJK (AM PM) | 14.58 | 1918 |
| AJK (AM FYM) | 13.22 | 1739 |
| AJK (AM SS) | 12.58 | 1655 |
| AJK (AM B) | 15.24 | 2005 |
| AJK (AM C) | 16.11 | 2120 |

Table 3. Atrazine Sorption Coefficient ($K_d$ & $K_{oc}$) for Attock Soil.

| Soils | Kd (L kg$^{-1}$) | $K_{oc}$ (L kg$^{-1}$) |
|---|---|---|
| Attock (UN AM) | 4.67 | 2748 |
| Attock (AM PM) | 6.41 | 3771 |
| Attock (AM FYM) | 5.20 | 3063 |
| Attock (AM SS) | 5.98 | 3520 |
| Attock (AM B) | 7.34 | 4322 |
| Attock (AM C) | 8.49 | 4999 |

UN AM = UN-Amended, AM PM = Amended Poultry Manure, AM FYM= Amended Farm Yard Manure, AM SS= Amended sewage sludge, AM B= Amended biochar, AM C= Amended Compost

## Discussion

Soil organic matter in soil influences the destiny of pesticides in soil. As the amount of SOM increases the sorption also increases. However literature revealed that the nature and type of organic materials also affect the rate of sorption. Size and types of different forms of organic materials had different impact on sorption. This experiment was performed to check the effects of sources organic amendments on the atrazine behaviour



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as sewage sludge, farm yard manure, poultry manure, biochar and compost were used. From the findings of present study it was noted that there was a positive connection between sorption and addition of exogenous organic carbon to the soil. Biochar and compost had greater influence on sorption as compared to the other three forms used because of their stable and small sizes. The results of current study were in line with the findings of Pérez-Lucas *et al.* (2021) who mentioned that consequences of exogenous sources of organic carbon on the behaviour of pesticides in soils because the efficacy of some pesticides and their perseverance and potential as environmental contaminants depend on their retention and degradation on soil constituents. Similarly, Worrall *et al.* (2001) stated that destiny of pesticides possibly influenced by organic amendments as addition of these alterations to soil upsurges organic matter which is considered the major component for retention of atrazine in soil.

Also the Findings of current study were in line with the observations of Macdonald *et al.* (2014) who mentioned that biochar addition to the soils increased the sorption of atrazine from 7 to 92 $Lkg^{-1}$. Zhang *et al.* (2021) also observed a positive relation of biochar to soils. They demonstrated that accumulation of biochar increased the sorption of atrazine from 200 to 2300% than without any addition of biochar. Yu *et al.* (2010) also detected that sorption of pesticides was enhanced in soil added with different biochars. This study shown in Table 4 and 5 demonstrated that application of compost on soil had great influence of sorption capacity of soil. Maximum value of $K_d$ was observed in both soils added with compost as compared to all others treatments. $K_d$ values for amended with compost in AJK and Attock soil were 16.11 $LKg^{-1}$ and 8.49 $LKg^{-1}$, respectively, whereas in un-amended those were 11.69 $LKg^{-1}$ and 4.67 $LKg^{-1}$, respectively. It was shown from data that maximum influence of compost application was measured in other all treatments which was due to nature of organic matter present in compost and its fine size such as humus which provides maximum sites for the sorption of atrazine on soil.

Results of the current study are in according to the findings of Sluszny *et al.* (1999) who reported reduction in water-soluble fraction of organic matter in compost and it increases humus level in soil which is responsible for the retention of atrazine. Houot *et al.* (2001) also mentioned that accumulation of two types of composted manures increases the sorption of atrazine in soil as compost application is answerable for the manufacture of huge amounts of humus. Atrazine transformation in soil after compost addition during long term laboratory incubations, they experienced that compost addition to soil generally inclined to decline atrazine mineralization and favored the stabilization of the corresponding residues but with most of them being un-extractable as it formed bound residues.

Results of the current study demonstrated that addition to soil increased sorption in both soils. The $K_d$ of un-amended soils were 11.69 $Lkg^{-1}$ and 4.67 $Lkg^{-1}$ for AJK and Attock soils respectively. When these soils were amended with poultry manure, a substantial rise in $K_d$ was observed in both soils as AJK and Attock amended soil has $K_d$ 14.58 $Lkg^{-1}$ and 6.41 $Lkg^{-1}$, respectively.

Findings were in accordance to the observations of Francou *et al.* (2005) who demonstrated that addition organic matter increases the sorption of pesticides. This was also in similar to the findings of Ahmad *et al.* (2001) who reported the significant influence OM on the sorption of atrazine in different soils as the poultry manure rich in total carbon content and addition to soil rises the organic carbon contents of soil which surges the sorption capacity of soils.

Subbaiah (2019) reported that carbon-rich organic materials such as FYM and poultry manure provide soluble and insoluble organic matter that possibly will change herbicide sorption as they contain hydrophobic compounds which also increases the sorption



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capacity because the materials provides more bindings sites for the adsorption of herbicides on its surface (Iqbal *et al.*, 2022). Results were also similar to the findings of Jaswal *et al.* (2022) who reported that accumulation of organic manure to soils may lead to addition of functional groups which provide surfaces for the adsorption these pesticides. It was also related to the findings of Basu and Rao (2020) who also stated that FYM application to soils disturb the destiny of herbicides that is controlled by many factors like volatilization, degradation, sorption and desorption. In low solubility compounds, interactions between dissolving organic matter and herbicides occur.

## Conclusion

Pesticides attribute in soils in influenced by SOM, types and amount of clay contents and also by soil pH. The current study showed that soils of AJK had more sorption capacity of atrazine as compared to Attock soil. The difference in their sorption capacity of atrazine in the soils (AJK & Attock) was due to differences in their mineralogy and soil chemicals properties including soil pH, SOM which are considered the key factors for the atrazine sorption. Therefore, it was concluded from these findings that rate and amount of pesticides applied to soils must be regarded soil chemicals and physicals properties which effect its movement into soils. The study also suggests that in order to reduce the soil amendments with the organic sources play as key role.

## Conflict of Interest

The authors have not declared any conflict of interest.

## Authors Contributions

All the authors contributed equally in the manuscript.

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