UDK 632.954:632.95.024
Pregledni rad – Review paper

Datum prijema: 29.9.2023.
Datum odobrenja: 16.7.2024.
Doi broj: 10.5937/33ah-46829

Paraquat and other dessicants and bleaching herbicides – their influence on weeds, crops and human and animal health

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SUMMARY

The paper provides an overview of various aspects of desiccants and bleaching herbicides, from different chemical groups, which have in common the drying and bleaching of leaf mass. In weed plants, they can act as total or selective herbicides, and be applied in the mature stages of the crop, where by drying the leaf mass they promote the maturation of some important crops such as sunflower, soybeans, and potatoes. More or less, all these various herbicides are highly or significantly toxic, which greatly complicates their application. In the EU, there was an initiative to ban some of them (e.g. paraquat). However, despite their significant toxicity, we believe they should be considered as an alternative to some other herbicides.
Keywords: desiccants, bipyridyl herbicides, paraquat, weeds, crop plants, toxicity.

INTRODUCTION

Weeds have been encountered by humans since the beginning of the development of agriculture, simply because the selection of cultivated plants is most often carried out by cross-breeding (Quarrie, 1997). This process enhances the quantitative traits of the cultivated plant, such as crop yield (economically the most important property of cultivated plants), but significantly reduces the competitive ability of cultivated plants against native species that grow around them, which are condidered weeds by definition. Historically, humans have always solved that problem manually, by weeding and other methods of physically removing weeds, so the rapid development of plant physiology and organic chemistry in the first half of the 20th century created the preconditions for a completely different way of solving this important problem. Namely, this knowledge led to the fact that in the years before the Second World War, weeds could be selectively removed chemically, without major negative consequences of those compounds on the health and yield of cultivated plants. After World War II, these promising findings led to systematic research into the synthesis of various (primarily organic) compounds and their testing in terms of weed removal efficiency and selectivity, i.e., the absence of significant negative effects of those phytotoxic compounds on cultivated plants (Corbet et al., 1984; Percival and Baker, 1991). Such an approach soon proved to be promising because while routine application of herbicides (as these phytotoxic compounds are called) leads to a reduction in crop yield due to weed competition of 8-15%, without the use of herbicides the total yield loss can reach up to 50% of the potential crop yield (Percival and Baker, 1991). The introduction into practice of a new agrotechnical measure, chemical crop protection, in addition to its benefits, also led to new unknowns and problems, one of which is the resistance of weeds to previously phytotoxic doses of herbicides, which testifies to the plastic variability of those native plants. This has become a serious problem, as the development of new phytotoxic formulations and herbicidal compounds is very expensive and demanding, while the profit can be small, depending on the economic expediency of using them. Additionally, there is the issue of a rapid development of weed resistance to some classes of herbicides (Percival and Baker, 1991; Travlos et al., 2020). This then entails the need for a more careful study of herbicidal effects, both on plants and on other living organisms and ecosystems, due to health and ecological concerns.

EFFECTS OF PARAQUAT AND OTHER DESICCANTS ON WEEDS

Paraquat (also known as methyl viologen) and its related compound diquat, as bipyridyl molecules (Averina et al., 1991), lead to the desiccation of weeds and partially controlled desiccation of crop leaf mass (for faster ripening), inhibiting the electron transport in Photosystem I (PSI) (Corbet et al., 1984). This is achieved when those electrons, instead of the usual photosynthetic acceptors (FNRS: feredoxine-NADPH oxydo-reduktase; Figure 1), are transferred to oxygen. This transfer occurs through Mehler's reactions or the water-water cycle (Kleczkowski, 1993; Janjić et al., 1994; Asada, 2000) producing reactive oxygen species, that disrupt cellular metabolism, leading to the drying and death of plant foliage.

In addition to the well-known bipyridyl herbicides with desiccation properties, and due to the development of resistance to bipyridyls (Streller et al., 1994; Weaver et al., 2004; Qin et al., 2004; Asaduzzaman et al., 2022; Farago et al., 2022), naturally occurring or due to genetic modification, mainly due to the increased content of antioxidant small compounds (e.g. polyamines) or antioxidant enzymes (which inactivate reactive oxygen species, thereby preventing phytotoxic processes associated with bipyridyls), a new type of desiccant that induces the Mehler reaction (s.c. water-water cycle) has recently been developed (Gerwiuck et al., 1997).

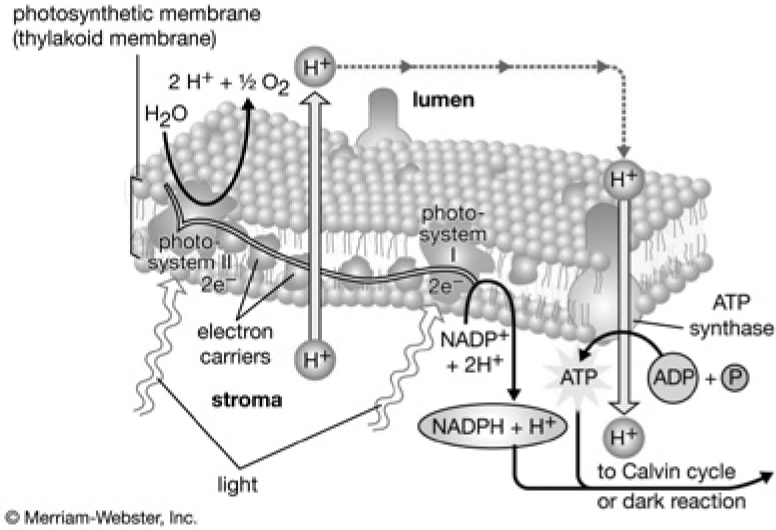

Figure 1. The transport of electrons in the light phase of photosynthesis
Slika 1. Transport elektrona u svetloj fazi fotosinteze

In addition to bipyridyls (as PSI inhibitors), which induce Mehler's reaction, there is another type of desiccant herbicides, which at the same time lead to leaf bleaching, indicative of increased reactive oxygen species production. These herbicides primarily function by inhibiting one of the pathways in the biosynthesis of chlorophyll or carotenoids (Sherman et al., 1991; Jacobs et al., 1991; Averina et al., 1991), targeting enzymes such as phytoene desaturase and protoporphyrinogen oxidase. These are diethyl ether, pyridazinone, and furanone type herbicides, as well as amitrole (Corbet et al., 1984; Percival and Baker, 1991). Such an approach, due to the appearance of weeds resistant to bipyridyl, influenced by the emergence of new desiccant herbicides (Ison et al., 2022), such as Voraxor®, a combination of the herbicides trifludimoxazin and saflufenacil: Tirexor® + Kixor®. However, it should be mentioned that paraquat, although generally considered a contact herbicide, has been observed to be taken up and transported through plants by the same pathways as polyamines (Fujita and Shinozaki, 2021), which indicates possibly new types of weed resistance to this and other desiccant herbicides (Farago et al., 2022; Lyu et al., 2022). In addition to other treatment practices, the efficacy of paraquat and other desiccants can be enhanced by encapsulation processes (e.g. with chitosan), which prolongs the otherwise short-term contact effect of this herbicide (Kurniadie et al., 2022).

EFFECTS OF PARAQUAT AND OTHER DESICCANTS ON CROP PLANTS

Although the application of herbicides is directed primarily against the harmful effects of weeds on crops, it is essential to also examine their impact on the growth, development, and yield quality and quantity of cultivated plants. This is importand due to the phytotoxic effects of these compounds on crops and possible residues they may leave in the harvested produce, thus entering the food and feed supply as toxic compounds. Therefore, studying the effects of desiccant herbicides is crucial, especially those classified as total herbicides (bipyridyls, amitrol), due to their persistence in both plant products and the soil where they are applied. All in all, it is necessary to distinguish the phytotoxic from the health aspects of the application of desiccant herbicides, when they are used in cultivated plants. The mode of action of desiccant herbicides on these plants varies widely, for example, bipyridyl herbicides such as paraquat, diquat and some related compounds, have many different effects on crop plants. In addition to inducing the production of reactive oxygen species (Asada, 2000), which disrupt cellular metabolism and lead to the decline of crop leaf mass (beneficial in the case of ripening of some crops, such as sunflower, soybean, potato), this group of herbicides causes pheophytinization, i.e. the loss of Mg ions from chlorophyll molecules. This process is another factor in the inhibition of photosynthesis and other metabolic processes and leads to an even greater production of reactive oxygen species (Averina et al., 1991). A series of studies by Nikolić et al. confirmed the pheophytinization of chlorophyll under the effects of diquat (Nikolić and Janjić, 1995; Nikolić et al., 1996a, 1996b; Nikolić, 1997; Pavlović et al., 2014). Additionally, significant changes in photosynthetic membrane proteins were observed under the effects of diquat (Milivojević and Nikolić, 1998, 1999; Milivojević et al., 1997), which were mediated by far-red light (Tables 1 and 2) (Milivojević and Nikolić, 1998). This light is primarily adopted through PSI, the main site of action of bipyridyl herbicides (Ashton and Crafts, 1981).

As indicated in the previous chapter, plant resistance to bipyridyl herbicides can be achieved through enzymatic antioxidant systems or through small molecules with antioxidant properties. This topic was investigated in detail on cultivated plants, and it was found that resistance to bipyridyl herbicides is increased by the action of small molecules (Hart et al., 1993; Donahue et al., 1997; Fujita and Shinozaki, 2021), which probably induce complexation of ions of transitional metals (Fe, Cu, Zn, etc.), as well-known promoters of the Mehler reaction (Chang and Kao, 1997). In addition to the effect on transition metals as mediators of the Mehler reaction, increased resistance to bipyridyl herbicides in cultivated plants is also possible through antioxidant enzyme systems (Kuk et al., 2006; Wang et al., 2021). Since the induction of the production of reactive oxygen species on a small scale has a favorable effect on plant metabolism, it was considered, as in the case of some other types of herbicides (Ashton and Crafts, 1981), that treating crop plants with small doses of bipyridyls might induce the hormesis

Table 1. Effects of diquat in the dark or under “white light” (WL) or far-red radiation (FR) during different time intervals (0-48h) on ratios of chlorophylls (Chl) and carotenoids (Car) in primary leaves of soybean and maize
Tabela 1. Efekati dikvata u mraku ili pod belom ili daleko-crvenom svetlošću tokom različitih vremenskih intervala (0-48h) na odnos hlorofila (Chl) i karotenoida (Car) u listovima soje i kukuruza

Crop/Usev	**Soybean/Soja**						**Maize/Kukuruz**									
Parameter Parametar	**Chl *alb***			**Chl a/Car**			**Chl *alb***					**Chl a/Car**				
Treatment Tretman	**0h**	**24h**	**48h**	**0h**	**24h**	**48h**	**5h**	**12h**	**18h**	**36h**	**48h**	**5h**	**12h**	**18h**	**36h**	**48h**
WL	2.1	2.3	2.4	5.3	5.6	4.6	2.6	2.5	2.3	2.5	n.m.	4.9	4.8	4.9	5.5	15.3
WL+ diquat	2.5	2.0	1.5	4.4	6.6	8.9	2.6	2.6	2.3	1.5	0.8	4.7	5.4	6.1	9.8	35.8
FR	2.2	2.1	1.7	3.4	3.4	3.9	2.6	2.4	2.4	2.1	2.1	4.9	4.6	4.9	4.6	4.9
FR+ diquat	2.1	2.1	1.5	3.5	4.4	6.9	2.6	2.4	1.5	1.9	0.9	5.2	4.6	15.8	6.8	11.6
Dark	2.1	2.3	2.6	5.3	4.1	4.2	2.8	2.7	2.5	1.6	1.2	5.0	4.7	5.2	5.7	6.0
Dark + diquat	2.3	2.4	2.1	4.1	4.1	5.1	2.7	2.6	2.4	1.4	1.0	4.9	3.6	5.7	13.8	10.7
LSD o.os		0.07			0.09				0.24					2.13		

n.m.: not measured. According to: Milivojević and Nikolić (1998)

Table 2. Effects of diquat in the dark or under “white light” (WL) or far-red radiation (FR) during different time intervals (0-24h) on polypeptides of photosynthetic reaction centres (RC) and light-harvesting complexes (LHC) of photosystem PSI and PSII of soybean and maize chloroplast thylakoids
Tabela 2. Efekati dikvata u mraku ili pod belom ili daleko-crvenom svetlošću tokom različitih vremenskih intervala (0-48h) na polipeptide fotosintetskih reakcionih centara (RC) i kompleksa za prikupljanje svetlosti (LHC) PSI i FSII tilakoida hloroplasta soje i kukuruza

Crop/Usev	**Soybean/Soja**										**Maize/Kukuruz**									
Parameter Parametar	**PS_1**		**PS_2**		**LHC**		**PS_2/PS_1**		**LHC/PS_2**		**PS_1**		**PS_2**		**LHC**		**PS_2/PS_1**		**LHC/PS_2**	
Treatment Tretman	**0h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**		**5 h→ 24 h**	
WL	7.1	3.0	23.2	38.6	35.4	34.3	3.3	1.8	1.7	1.0	6.9	5.2	22.2	33.8	36.7	29.8	3.2	6.5	1.7	0.8
WL+ diquat	4.0	2.6	19.1	25.4	40.7	43.7	4.8	9.8	2.1	1.7	1.9	4.0	24.7	33.6	42.6	25.6	13.0	8.4	1.7	0.8
FR	5.3	2.6	24.6	23.8	23.9	43.7	4.6	9.2	1.0	1.8	2.9	5.8	28.4	25.7	37.1	27.9	9.8	4.4	1.3	1.1
FR+ diquat	3.5	2.0	12.9	15.0	50.7	34.3	3.7	7.5	3.9	1.4	1.2	3.6	21.2	23.4	45.9	23.4	17.7	6.5	2.2	1.0
Dark	3.5	3.1	19.9	22.0	39.0	46.3	5.5	8.7	2.0	1.7	1.5	4.0	25.0	26.3	35.8	30.9	16.7	10.0	1.4	1.2
Dark + diquat	4.9	1.8	34.9	30.0	34.9	37.0	4.7	16.7	1.5	1.2	n.m.	2.6	45.0	32.0	36.3	25.1	45.0	12.3	0.8	0.8
LSD o.os	0.8		10.5		9.3		1.5		1.2		1.1		5.1		8.5		7.6		0.4	

n.m.: not measured. According to: Milivojević and Nikolić (1998)

effect, thereby increasing plant metabolism, which would theoretically have a favorable effect on crop yield. However, these attempts did not give sufficiently promising results (Ferrari et al., 2021), although similar trials have shown favorable results in the model plant of *Arabidopsis thaliana* (Farago et al., 2022). The effects of some other types of desiccant herbicides, lead to leaf bleaching at the same time, indicating an increased production of reactive oxygen species, with mechanisms of action based on the inhibition of one of the pathways in the biosynthesis of chlorophyll or carotenoids. In this case, we can observe the binding of these herbicides to enzymes such as phytoene desaturase and protoporphyrinogen oxidase (Dalla Vekia et al., 2001; di Baccio et al., 2001), which leads to their inhibition, and thus to the increased production of reactive oxygen species. However, there is a physiological difference in the degree of binding of these herbicides to the abovementioned enzymes, which may underline the potential resistance of cultivated plants to their effect (Sherman et al., 1991).

EFFECTS OF PARAQUAT AND OTHER DESICCANTS ON HUMAN AND ANIMAL HEALTH

Paraquat, partly diquat, and other desiccants are highly toxic substances. Paraquat is one of the most poisonous and according to the nomenclature applied in Serbia, classified in the 1st group of poisons (the highest toxicity category). This is partly due to the fact that paraquat induces the production of reactive oxygen species, which disrupt metabolism (Brown and Sejther, 1990) and damage human, animal and plant cells. Additionally, paraquat's high volatility can cause eye damage, and diquat can penetrate the skin. Consequently, bipyridyl compounds can easily enter into the body of humans and animals, through means other than inhalation or ingestion of contaminated food or liquids. Desiccant herbicides from other chemical groups mostly cause poisoning by ingestion via contaminated food or liquids, or when they are inadequately applied in the field. Nevertheless, unlike bipyridyls, which are highly toxic, but with a short-term effect due to rapid inactivation, desiccants from other chemical groups are persistent in the environment. This persistence can lead to their accumulation and prolonged toxicity to humans and animals. Detoxification and relief of bipyridyl poisoning symptoms are performed in different ways, either by removing or inactivating transition metal ions (Krall et al., 1991; Sato, 1991), which act as cofactors with bipyridyl in the production of reactive oxygen species, or by using natural (Palipoch et al., 2022; Chen et al., 2023) or synthetic (Li et al., 2021) drugs. The consequences of intoxication with bipyridyl herbicides are severe, causing damage to the eyes, lungs, liver, and kidneys, and potentially leading to long-term symptoms, causing Alzheimer's disease (Marshall and Prior, 2022). That is why, at the first instance of the Court of the European Union, Sweden, with the support of Denmark, Finland and Austria, and against the European Commission, launched an initiative to cancel the license for the use of paraquat as a plant protection product, due to its health risks (Anonymus, 2007). The EU court supported this initiative at that decision-making level.

OTHER ASPECTS RELATING TO PARAQUAT AND OTHER DESICCANTS

Because of these health risks, sophisticated analytical techniques have been developed for the detection of bipyridyl herbicide residues in various human, animal, and plant tissues (Hao et al., 2013; Tsao et al., 2016; Guo et al., 2023). Additionally, various techniques are being considered for the fastest possible inactivation of these highly toxic substances in the event of soil or water contamination (Inthama et al., 2021; Hammami et al., 2022).

CONCLUSIONS AND FUTURE PERSPECTIVES ABOUT THE EFFECTS OF PARAQUAT AND OTHER DESICCANTS IN AGRICULTURE AND RELATED AREAS

Bipyridyl herbicides, along with other desiccants, have shown high efficiency, either for the complete removal of weed vegetation (total herbicides: paraquat, diquat, amitrole), or for its selective removal (e.g. diethyl ether, pyridazinone, and furanone-type herbicides). However, their high toxicity (paraquat) and persistence in the environment increasingly raise concerns, leading to restrictions and even bans on their use. But certainly, despite these restrictions, their high efficiency in removing weeds cannot be ignored. Developed countries, similar to the case of some organochlorine insecticides (e.g. DDT), which are known for their high persistency and a proven harmful effect on the environment, still maintain stocks of these substances for emergency use. This is because having such options available can be crucial for sudden, critical needs. We believe that considering all the limitations in the application of these types of pesticides, our country must also develop a strategy for responding to possible palliative situations, even if it involves the use of these highly toxic herbicides.

ACNOWLEDGEMENT

The research was funded by the Ministry of Science of the Republic of Serbia, under Contracts No.: 451-03-66/2024-03/200010 and others.

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Parakvat i drugi herbicidi – desikanti i izbeljivači – njihov uticaj na korove, useve i zdravlje ljudi i životinja

REZIME

U radu je dat pregled različitih aspekata desikanata i izbeljivačkih herbicida, iz različitih hemijskih grupa, koji imaju zajedničku osobinu isušivanja i izbeljivanja lisne mase. U korovskim biljkama mogu delovati kao totalni ili selektivni herbicidi, a primenjeni u zrelim fazama useva, isušivanjem lisne mase promovišu sazrevanje nekih važnih useva (suncokret, soja, krompir). Većinu ovih herbicida karakteriše visoka ili znatna toksičnost, što znatno komplikuje njihovu primenu. U Evropskoj Uniji došlo je do inicijative za zabranu nekih od ovih aktivnih supstanci (npr. parakvat). Međutim, uprkos njihovoj značajnoj toksičnosti, verujemo da bi ih trebalo razmotriti kao alternativu nekim drugim herbicidima.

Ključne reči: desikanti, bipiridil herbicidi, parakvat, korovi, usevi, toksičnost.