Risk monitoring and assessment of plant growth regulators residues in fruits from Shandong Province, P. R. China

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Citation

Jiang Wei., Lan Feng., Zhou Xianxue., Li Xiaoliang., Liu Chuande., Wang Zhixin., Lu Zeqi., Duan Xiaona., Yao Jie., Wang Chunxiao., & Liu Xuan. (2020). Risk monitoring and assessment of plant growth regulators residues in fruits from Shandong Province, P. R. China. *Rastenievadni nauki, 57(3)* 17-22

Abstract

The residue and risk level of plant growth regulator in main producing areas of Shandong were clarified. From 2017 to 2018, 260 samples of cherry, grape, pear, jujube, blueberry and watermelon were collected from Shandong. 20 plant growth regulators were analyzed using liquid chromatography tandem mass spectrometry. Chronic dietary intake risk of plant growth regulators in fruits was evaluated by % Acute Reference Dose (% ARfD). A total of four plant growth regulators including abscisic acid (ABA), compound sodium nitrophenolate (CSN), indole-3-acetic acid (IAA) and chlormequat chloride (CCC) were detected, and the maximum residual amount were 2.49, 1.60, 0.038 and 0.048 mg/kg, respectively. Risk assessment results showed that the acute dietary intake risk (% ARfD) of detected plant growth regulators was 0.2 to 14.5. The acute dietary risk of plant growth regulator in fruit was low.

Keywords: fruit; plant growth regulators; residues; risk assessment

Plant growth regulators (PGR) has been widely used as a technical measure of high efficient and high yield agriculture. But, in recent years, media has been rendering seedless grapes, top flower with cucumber are using contraceptives, hollow strawberry is from use of hormones, such as public opinion, made a great impact to the relevant industry. Therefore, to find out the residual status and risk level of the main fruit plant growth regulators, and to clarify the industrial development needs and regulatory focus, it has guiding significance to guide consumption, industrial development and quality safety.

At present, Li Qian et al. (2016) carried out monitoring and analysis of 6 kinds of PGR of fruits such as Gibberellin in Daxing District Beijing. Jiang Nan et al. (2015) and Luq et al. (2014) monitored the residues of plant growth regulators in fruits in 9 areas of Jilin Province, and analyzed the results to provide scientific basis for related food safety policies and regulations. The application and residue of plant growth regulators are different because of different climate conditions, production management and level. The risk monitoring and assessment of 20 kinds of PGR commonly used in fruit production areas in Shandong Province were carried out, to lay a foundation for promoting the registration of PGR and the revision of maximum residue limit and detection standard, guiding consumption and promoting the healthy development of the industry.

MATERIALS AND METHODS

According to the fruit yield planting area and brand influence of various regions in Shandong, from 2017 to 2018 in the main fruit producing areas of Shandong, through on-the-spot investigation, a total of 260 samples of 6 kind of fruits were collected from facilities with high social concern, such as Sweet Cherry planted in green house (Yantai, Tai'an, Weifang), grape (Yantai, Qingdao), watermelon (Weifang, Yantai), Jujube (Binzhou, Dezhou, Tai'an), blueberry (Weihai, Yantai) and pear (Yantai).

The detection method is based on the literature report method, and 20 kind of plant growth regulators were test in this paper, including 2,4-Dichlorophenoxyacetic acid, 4-Chlorophenoxy acetic acid, Paclobutrazol, Uniconazole, Chlormequat chloride, Mepiquat chloride, Forchlorfenuron, Thidiazuron, Naphthaleneacetic acid, Gibberellin, Brassinolide, Sodium nitrophenolate, Indole-3-acetic acid, Diethyl aminoethyl hexanoate, 6-Benzylaminopurine, Abscisic acid, Daminozide, Clopyralid, Trinexapac-Ethyl, 2,3,5-Triiodobenzoic acid.

The risk of acute dietary intake (% ARfD) was calculated using the JMPR recommended formula (Gao et al., 2007; Lan et al., 2015). Formula (1) is suitable for the case where the weight of a single fruit is less than 25g, formula (2) is suitable for the case where the weight of a single fruit is more than 25g and less than that of a large portion, and formula (3) is suitable for the case where the weight of a single fruit is more than that of a large portion.

$$\% ARfD = \frac{LP \times HR}{bw \times ARfD} \times 100 \quad (1)$$

$$\% ARfD = \frac{U \times HR \times v + (LP - U) \times HR}{bw \times ARfD} \times 100 \quad (2)$$

$$\% ARfD = \frac{LP \times HR \times v}{bw \times ARfD} \times 100 \quad (3)$$

Of (1)(2)(3), % ARfD is acute dietary risk; LP is large portion, measuring unit is kg; HR is Maximum Residue Limit, measuring unit is mg/kg; U is the weight of single fruit, measuring unit is kg; v is the variation factor, and the value is 3; bw is body weight (kg); ARfD is Acute Reference Dose (mg/kg).

RESULTS AND DISCUSSION

A total of 6 PGR residues were detected from 6 kinds of fruits in the main production area of Shandong Province (Figure 1). The detection rates from high to low were Abscisic acid (90%), Sodium 2-nitrophenoxide(62%), 4-nitrophenol sodium salt dehydrate (62%), 2-Methoxy-5-nitrophenol sodium salt (6%), Chlormequat chloride (3%), Indole-3-acetic acid (1%).

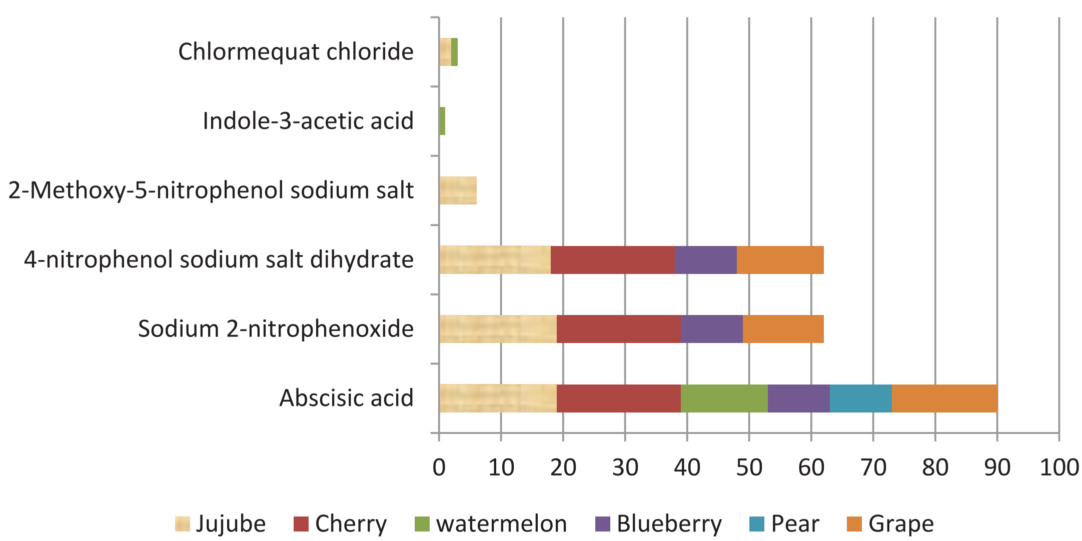

Figure 1. Detection rate of plant growth regulators in fruits

The PGR species were found in cherry, grape and blueberry, including Abscisic acid, Sodium 2-nitrophenoxide (62%)and 4-nitrophenol sodium salt dihydrate. Abscisic acid, Indole-3-acetic acid and Chlormequat chloride were mainly detected in watermelon. Abscisic acid, Sodium 2-nitrophenoxide, 4-nitrophenol sodium salt dihydrate, 2-Methoxy-5-nitrophenol sodium salt and Chlormequat chloride were mainly detected in jujube. Only Abscisic acid was detected in pears.

Abscisic acid was detected in 6 kinds of fruits, and the highest was detected in blueberry (2.49 mg/kg) (Table 1).

Sodium 2-nitrophenoxide and 4-nitrophenol sodium salt dihydrate were detected in grape, jujube and blueberry, but their residues were higher in Jujube. 2-Methoxy-5-nitrophenol sodium salt was detected only in Jujube, Indole-3-acetic acid was detected only in watermelon, and Chlormequat chloride was detected in watermelon and jujube. 2,4-Dichlorophenoxyacetic acid, 4-Chlorophenoxy acetic acid, Paclobutrazol, Uniconazole, mepiquat chloride, Forchlorfenuron, Thidiazuron, Naphthaleneacetic acid, Gibberellin, Brassinolide, Sodium nitrophenolate, Diethyl aminoethyl hexanoate, 6-Benzylaminopurine, Daminozide, Clopyralid, Trinexapac-Ethyl, 2,3,5-Triiodobenzoic acid were not detected in 6 kinds of fruits.

Abscisic acid, Indole-3-acetic acid, Sodium 2-nitrophenoxide, 4-nitrophenol sodium salt dihydrat, 2-Methoxy-5-nitrophenol sodium salt and Chlormequat chloride were detected in 6 kinds of fruits in the main production area of Shandong Province. Abscisic acid and Indole-3-acetic are plant endogenous hormones, and many countries do not have maximum residue limits (MRLS), so there is no need to assess their dietary intake risk. The MRL value of Chlormequat chloride is only established in food crops. Sodium 2-nitrophenoxide, 4-nitrophenol sodium salt dihydrat, 2-Methoxy-5-nitrophenol sodium salt are collectively referred to as Compound Sodium Nitrophenolate.

The temporary maximum residue limits of Compound Sodium Nitrophenolate in wheat, soybean, tomato and orange were also established in China (Maly et al., 2013; Song et al., 2017). In this study, the MRL value could not be used to evaluate the risk of Chlormequat chloride and Compound Sodium Nitrophenolate deficiency in 6 fruits. The JMPR published a formula to calculate the risk of acute dietary intake, by which the risk of Chlormequat chloride and Compound Sodium Nitrophenolate in fruits was evaluated. The results are shown in Table 2.

The acute dietary intake risk % ARfD of Chlormequat chloride and Compound Sodium Nitrophenolate in 6 kinds of fruits was between 0.2 and 14.5, which was much less than 100, indicating that the acute dietary intake risk of Chlormequat chloride and Compound Sodium Nitrophenolate in 6 kinds of fruits was lower.

There are currently no plant growth regulators registered for sweet cherries and blueberries. More than 100 plant growth regulators have been registered in grape, Jujube, watermelon and pear, of which 72 have been registered in grapes. Through

Table 1. Kinds of plant growth regulators (PGR) residues detected in fruits

PGR	LOD (mg/kg)	Residual quantity (mg/kg)					
		Cherry	Grape	Jujube	Blueberry	Watermelon	Pear
Abscisic acid	0.002	0.013~0.403	ND~1.45	0.07~0.974	0.41~2.49	ND~0.346	0.082~0.651
Sodium 2-nitrophenoxide	0.003	0.017~0.111	ND~0.163	0.034~0.848	0.01~0.032	ND	ND
4-nitrophenol sodium salt dihydrate	0.003	0.023~0.078	ND~0.111	0.026~0.696	0.01~0.019	ND	ND
2-Methoxy-5-nitrophenol sodium salt	0.003	ND	ND	ND~0.056	ND	ND	ND
Indole-3-acetic acid	0.005	ND	ND	ND	ND	ND~0.038	ND
Chlormequat chloride	0.004	ND	ND	ND~0.02	ND	ND~0.048	ND

Table 2. Acute dietary intake risk of plant growth regulators in fruits

Fruit	Large portion / kg	Unit/kg	Risk factor	Maximum Residue Limit mg/kg	Acute Reference Dose (ARfD) mg/kg bw	Dietary intake risk, % ARfD
Cherry	0.713	0.012	Sodium 2-nitrophenoxide	0.111	0.045	2.8
			4-nitrophenol sodium salt dihydrate	0.078	0.045	2.0
Jujube	0.286	0.020	Sodium 2-nitrophenoxide	0.848	0.045	8.6
			4-nitrophenol sodium salt dihydrate	0.696	0.045	7.0
			2-Methoxy-5-nitrophenol sodium salt	0.056	0.045	0.6
			Chlormequat chloride	0.02	0.05	0.2
Grape	0.570	0.637	Sodium 2-nitrophenoxide	0.24	0.045	14.5
			4-nitrophenol sodium salt dihydrate	0.111	0.045	6.7
Watermelon	2.542	2.096	Chlormequat chloride	0.048	0.05	10.3
Blueberry	0.388	0.0018	Sodium 2-nitrophenoxide	0.032	0.045	0.4
			4-nitrophenol sodium salt dihydrate	0.019	0.045	0.3

field investigation, it was found that Compound Sodium Nitrophenolate was widely used in production, and its main function was to increase fruit setting rate, increase yield and improve quality. The results showed that the detection rate of Compound Sodium Nitrophenolate in grape, cherry, blueberry and jujube was over 60%. At present, Compound Sodium Nitrophenolate has been registered mainly in grain and vegetables, but not in fruits. According to the analysis results of verification and the actual production situation, it is suggested to register compound Sodium nitrophenolate on fruits.

The maximum residue limits (MRLS) of plant growth regulators (PGR) have not been established in pear and Jujube, but only one MRLS in Cherry, watermelon and blueberry and five MRLS in grape. Compared with the registered PGR, the established limits of plant growth regulators are insufficient. Based on the production practice and the research results, it is suggested that the standard limit values of Compound sodium nitrophenolate in 6 kinds of fruits, such as cherry, should be established.

In 2008, European Food Safety Authority studied the toxicological properties of Compound Sodium Nitrophenolate and found that it can be rapidly absorbed and widely distributed in the body, posing a risk of serious harm to the eyes. In 2015, European Food Safety Authority set the limit for Sodium 2-nitrophenoxide, 4-nitrophenol sodium salt dihydrat, 2-Methoxy-5-nitrophenol sodium salt in fruits, vegetables and grains at 0.03 mg/kg.

The temporary limit of Compound Sodium Nitrophenolate in wheat, soybean, tomato and orange in China is 0.1 mg / kg. In this study, the maximum residue of Compound Sodium Nitrophenolate was 1.60 mg / kg, which was much higher than the limit of 0.03 mg / kg set by European Food Safety Authority. Although the acute dietary intake risk of Compound Sodium Nitrophenolate is not high, due to its special toxicological characteristics, there is a risk of serious injury to the eyes, and a high residual level of Compound Sodium Nitrophenolate may have potential chronic dietary intake risk.

It is suggested that further study on toxicological characteristics and dietary intake risk of Compound Sodium Nitrophenolate should be carried out to provide technical support for establishing maximum residue limit standards, ensuring food safety and import and export trade. In addition, the detection methods of PGR residues are scattered, the pretreatment methods are not uniform, some PGR still need to be derived after detection, the detection steps are cumbersome. Some PGR tests lack standards. It is suggested that the development of multi-residue detection standard for PGR should be accelerated.

Since 2015, China has launched the risk assessment of quality and safety of plant growth regulators in agricultural products. Previous work has focused on mapping the use and residue levels of PGR in agricultural products, conducting a Exploratory research on the impact of PGR on the nutritional quality of agricultural products, and responding to safety issues of high public concern. Because of the use of compound PGR in the production and the use of different kinds of PGR in different growing stages of crops, there are many residues of PGR in agricultural products. In addition, PGR metabolites and PGR auxiliaries on the quality and safety of agricultural products should also attract the attention of relevant researchers. Some PGR metabolites and auxiliaries have been shown to be more toxic. Therefore, the future risk assessment of fruit PGR should focus on the risk of multi-residue combined exposure, the effects of PGR metabolites and pesticide additives on quality and safety.

CONCLUSIONS

From 2017 to 2018, 260 samples of sweet cherry, grape, pear, jujube, blueberry and watermelon were collected from Shandong province. 20 plant growth regulators were analyzed using liquid chromatography tandem mass spectrometry. Chronic dietary intake risk of plant growth regulators in fruits was evaluated by % ARfD. A total of four plant growth regulators including abscisic acid, compound sodium nitrophenolate, indole-3-acetic acid and chlormequat chloride were detected, and the detection rate of Abscisic acid and Compound Sodium Nitrophenolate was high. The risk assessment showed that the detected acute dietary exposure to PGR was low. In view of the actual production needs and monitoring results, it is suggested to register Compound Sodium Nitrophenolate in fruits, give priority to setting up the limit standard of Compound Sodium Nitrophenolate in fruits, and speed up the establishment of the multi-residue detection standard of PGR. At the same time, the risk assessment of PGR multi-residue combined exposure, PGR metabolites and pesticide auxiliaries on fruit quality and safety should be studied in the future.

ACKNOWLEDGMENT

Fund project: National Fruit Quality and Safety Risk Assessment (GJFP2018003), Agricultural product plant growth regulator use safety assessment (GJFP20180014), Shandong Academy of Agricultural Sciences and Technology Innovation. Project (CXGC2016B17).

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