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PLASTIC GREEN HOUSE CUCUMBER FRUITS POLLUTION WITH FUNGICIDES: (2) IN EGYPTIAN MARKETS

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ABSTRACT

Fresh cucumber fruits samples of plastic green house were marketed from eighteen localities representing twelve Egyptian Governorates. Residues of Diniconazole (Sumi-eight), Carboxin (Vitava x200), pyrazophos (Afugan), Fenarimol (Rubigan), and Metalaxyl (Radomil plus) were thoroughly investigated and determined on and in unwashed and washed fruits. Experimental results showed that the residues of Diniconazole and carboxin in samples of all different eighteen localities were either undetectable or loss than maximum residues limits (MRL), except which collected from El-Helmia El-Kadema and Modriet El-Tahrir where the both fungicides residues were more than MRL, the residues of pyrazophos in samples collected from ten locations were more than MRL, while those collected from El-Helmia El-Kadema, Embaba, Dokki, Dikerness, Zagazig, Kom Hamada, El-Arab, and Kena were either undetectable or less than MRL, also the residues of Fenarimol in samples collected from most locations were either undetectable or less than MRL, while those collected from El-Agamy, Benha, and Dikerness were more than MRL, and latter the residues of Metalaxyl in samples collected from most locations were either undetectable or lower than MRL, while those collected from Heliopilies and Rod El-Farag were more than MRL.

Therefore, unwashed and washed cucumber fruits samples collected from twelve Egyptian markets as follows: Heliopilies, Bab El-Loak, Rod El-Farag, El-Helmia El-Kadema, Giza, El-Agamy, Benha, Menouf, Dikerness, Modriet El-Tahrir, Fayed, and Minia were not safe for human consumption, while those collected from Embaba, Dokki, Zagazig, Kom Hamada, El-Arab, and Kena can be consumed safely by human.

INTRODUCTION

The widespread use of systemic and non-systemic fungicides during the Fungi decade in plastic green houses for the control of injurious fungi on vegetables crops reverted the importance of certain problems. It is assumed that some residues of such fungicides could detect in varying concentrations after their application such remaining residues could be hazard to the consumers health or the products quality. This hazards could be further enhanced in case of vegetable fruits which are usually consumed freshly e.g. consumed vegetable fruits freshly contaminated with fungicides residues, more than the allowable tolerance. Even after washing which is not always carried out or could be inefficient, higher levels of those residues.

The effect of commercial and preparative method as washing on levels of fungicides residues studied by different authors (Kepczynska 1989, El-Sheamy and Ramadan 1990, Koriem *et al.*, 1991, and Hussien *et al.,* 1996). The present study was conducted to determine some fungicides residues on and in unwashed and washed fresh cucumber fruits samples from different Egyptian markets after harvesting, and foliar spraying under

plastic green house conditions in Egypt. This research is recommend must be prevent foliar spraying enough interval before harvesting to avoid the hazards involved, this period need to particularly studied for each fungicides, and also fungicides residues must be determined to know if these residues are within the acceptable limits or not.

MATERIALS AND METHODS

Sampling for Fungicides residues analysis:

Plastic green house cucumber fruits samples were randomly collected during April, 2000 from eighteen Egyptian markets of different Governorate as follows: Cairo (Heliopiles, Bab El-Loak, Rod El-Farag, and El-Helmia El-Kadema), Giza (Giza, Embaba, and Dokki), Alexandria (El-Agamy), Qualubia (Benha), Menoufyia (Menouf), Dakahlia (Dikerness), Sharkia (Zagazig), Behera (Kom Hamada and Modriet El-Tahrir), Ismailia (Fayed), Port Said (El-Arab), Mini (Mallawi), and Kena (Kena).

Random fresh cucumber samples of about 2kg were collected from the markets, the representative samples was divided into two subsample (1kg), the first was left unwashed and the second washed by soaking for 3 minutes in running tap water followed by spraying for 2 minutes with tap water then air dried.

The experiments were carried out for determination of five fungicides residues as follows: Metalaxyl, Diniconazole, Carboxin, Pyrazophos, and Fenarimol on and in unwashed and washed samples of fresh cucumber fruits which were treatment by the programme from Ministry of Agriculture (Egypt) under plastic green house conditions according to the recommended rate with different fungicides (Pest Control programme of vegetables 1993 page, 91and pest of agriculture control programme 1997 page 98).

Extraction and Clean-up:

Metalaxyl residues was extracted and cleaned-up from cucumber fruits samples according to the method of David and Unwin (1981), and Diniconazole residues was extracted and cleaned-up according to the method of El-Bouze *et al.* (1998), while carboxin, pyrazophos, and fenarimol residues were extracted and cleaned-up according to the method of Macnell *et al*. (1975).

The R_f values were determined by TLC and results appearted that: Metalaxyl 0.67, Diniconazole 0.59, carboxin 0.71, pyrazophos 0.64, and fenarimol 0.58.

Determination:

The fungicides residues were determined by gas-liquid partition chromatography Instrument (GLC). Residues dissolved in methanol were then estimated using GC fitted with flame ionization Detector (FID), carrier gas flow rate 40 ml nitrogen/min., Capillary column, and Temperature programming (Table i) also GC chromatograms of these fungicides standards and Rt shown in fig (i).

Table (i): GLC conditions, for residue analysis of fungicides.

Conditions / Fungicides	Oven Temp	Init Value	Init Time	Rate Deg./ min	Final Value	Final Time (min)	Inj. Temp	Det. Temp	Atten -uat- ion	Rete ntion time
Diniconazole	199	199	1	5	225	4	269	269	16	3.30
Pyrazophos	225	225	1	5	259	8	269	269	2	3.80
Fenarimol	235	235	1	5	259	8	279	279	16	3.23
Carboxin	199	199	2	15	235	4	269	269	16	3.66
Metalaxyl	199	199	2	15	235	4	245	261	4	1.51

Table (ii): Average Rate of recoveries of tested compound

Fungicides	Recovery %
Diniconazole	86.25
Pyrazophos	89.20
Fenarimol	84.00
Carboxin	87.15
Metalaxyl	85.87

The same methods of extraction were followed using know concentrations of fortified samples of fungicides with each to obtain rate of recoveries (Table ii)

RESULTS AND DISCUSSION

The study of fungicides persistence and residual behaviour gives an idea about the pre-harvest interval that should pass following application and before marketing in order to minimize health hazards. On the other hand, many investigators emphasized the role of home and commercial preparations in removing the pesticides residues from agricultural products (Liska and Stademan 1969. Geisman 1975 and Celino and Magallona 1985, Besser *et al*., 1991).

The use of fungicides in protecting vegetable from fungi infestation has been recently increased therefore, the residue limits of such fungicides should be emphasized for human safety during consumption.

Fungicides exhibit plant growth regulating properties and these fungicides may be regarded as an activation because the metabolite possesses greater fungicidal activity than the parent compound. (Macdonald and Meyer 1998).

The fungicides were used one or more into cucumber plants grown under greenhouse. Abd El-Aziz (1998) found residues of used pesticides in cucumber fruits increased with increasing number of insecticidal application

Diniconazole residues :

Systemic and non-systemic fungicides have been used widely for controlling wide range of economically important diseases minaly in vegetables crops. Therefore enhanced use of these fungicides especially foliar spray, increased the environmental pollution.

As show in table (1) fresh cucumber fruits samples which marketed from Kom Hamada, Fayed, El-Arab, Minia, and Kena locations were free from any Diniconazole residues, while those marketed from other locations recorded Diniconazol residues varied from 0.15ppm in Embaba to 7.00ppm in Modriet El-Tahrir.

Table (1): Level of Diniconazole and Carboxin residues on and in unwashed and washed cucumber fruits (ppm)

Fungicides / Samples / Location	Diniconazole			Carboxin		
	Unwashed	Washed	% Loss by washing	Unwashed	Washed	% Loss by washing
Heliopiles	2.34	0.41	82.48	0.69	0.09	86.96
Bab El-Loak	0.40	0.06	85.00	0.74	0.10	86.49
Rod El- Farag	1.78	0.39	78.09	UND	UND	-
El-Helmia El-Kadema	3.31	0.81	75.53	4.91	1.19	75.76
Giza	0.27	0.03	88.89	0.40	0.05	87.50
Embaba	0.15	0.02	86.67	UND	UND	-
Dokki	1.60	0.34	78.75	0.55	0.08	85.45
Al-Agamy	0.92	0.20	78.26	1.46	0.34	76.71
Benha	0.24	0.05	79.17	1.61	0.42	73.91
Menouf	0.45	0.08	82.22	1.11	0.21	81.08
Dikerness	1.43	0.21	85.31	UND	UND	-
Zagazig	0.55	0.09	83.64	0.39	0.09	76.92
Modriet El-Tahrir	7.00	1.24	82.29	5.30	0.86	83.77
Kom Hamada	UND	UND	-	UND	UND	-
Fayed	UND	UND	-	0.21	0.04	80.95
El-Arab	UND	UND	-	3.67	0.48	86.92
Minia	UND	UND	-	UND	UND	-
Kena	UND	UND	-	UND	UND	-

UND = Undetectable

Washing by running tap water removed 75.53% to 88.89% of Diniconazole residues from contaminated samples. So this meaning, washing process gave higher decontamination of Diniconazole residues. Consequently, unwashed and washed cucumber samples which collected from eighteen different Egyptian markets were lower than the maximum residual limits (MRL) (0.5mg/kg for Diniconazole in cucumber fruits) by CAC 1996 and CAC 1997 except which collected from El-Helmia El-Kadema and Modriet El-Tahrir, where Diniconazole residues were more than MRL 0.81 and 1.24ppm, respectively.

The degradation of Diniconazole systemic fungicide was assumed to occur enzymically within the symplast of cucumber fruits. These results inagreement with these of Solel *et al*. (1973) who reported that a small but definite percentage of systemic fungicide applied to leaves is transport to roots and shoots, via the phloem.

Carboxin residues:

Data in table (1) indicated that fresh cucumber fruits samples which marketed from Rod El-Farag, Embaba, Dikerness, Kom Hamada, Minia, and

Kena localities were free from any carboxin residues, while those marketed from other localities recoded carboxin residues varied from 0.21ppm in Fayed to 5.30ppm in Modriet El-Tahrir.

Washing process removed 73.91% to 87.50% of carboxin residues from contaminated samples, this washing gave higher decontamination of carboxin residues from cucumber samples. After washing process, unwashed and washed cucumber samples which marketed from eighteen different markets were lower than MRL (0.5mg/kg for carboxin in cucumber fruits) except which marketed from El-Helmia El-Kadema and Modriet El-Tahrir where the residues of carboxin were more than MRL 1.19 and 0.86ppm, respectively.

Uptake and translocation of carboxin in cucumber plant, root uptake and translocation to stem and leaves from both an aqueous solution and drenched soil and last to fruits. These results are line with those reported by Solel and Pinkas (1972).

Pyrazophos residues:

In table (2) the data indicated that fresh cucumber fruits which marketed from seven different markets as follows: El-Helmia El-Kadema, Dokki, Dikerness, Zagazeig, Kom Hamada, El-Arab, and Kena were free from any pyrazophos residues, while those marketed from eleven locations were contaminated with this fungicide, and the residues were varied from 0.13ppm to 5.20ppm in Embaba and Bab El-Loak, respectively.

Washing process removed 51.64% to 61.54% of pyrazophos residues from contaminated samples. Therefore, unwashed and washed cucumber samples which marketed from ten different regions were exceeded the MRL (0.1mg/kg for pyrazophos in cucumber fruits), while cucumber samples which marketed from eight different regions as follows: El Helmia El-Kadema, Embaba, Dokki, Dikerness, Zagazig, Kom Hamada, El-Arab, and Kena were either undetectable or lower than MRL.

Fenarimol residues:

Results in table (2) indicated that samples collected from Bab El-Loak, Embaba, Zagazig, Modriet El-Tahrir, Kom Hamada, El-Arab, Minia, and Kena locations were free from any fenarimol residues, while those collected from other locations were contaminated with this fungicide. Residues varied from 0.05ppm to 5.80ppm in Rod El-Farag and Dikerness. Washing caused reduction of the obtained residues by 60.00% to 87.21% and gave higher decontamination of fenarimol residues. After washing process, cucumber samples which marketed from all eighteen different markets were lower than MRL (0.5mg/kg for fenarimol in cucumber fruits) except which marketed from El-Agamy, Benha, and Dikerness where residues of fenarimol were 0.73, 0.57, and 2.15ppm respectively. The above results are in line with those reported by Nagagmi, H. (1997) who studied that the amounts of the fungicide residues in the strawberry samples on the market in Nara, Japan.

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Luke *et al*. (1988) surveyed 342 strawberry samples on the market in California, USA and reported that vinclozolin fungicide residues more than 0.1ppm and analysed 3 fungicides (fenarimol, myclobutanil, triadimefon) and found that no residues was detected from 342 strawberry samples in California, USA.

Camoni *et al*. (1993) surveyed fenarimol residue in Italy, and reported that no residue was detected in 215 strawberry samples, and Neidert *et al.* (1994) analysis fenarimol and triadimefon, and reported that no residue was detected from 48 strawberry samples on the market in Canada.

Also, these obtained results are in agreement with those of cabras *et al*. (1985) as they reported that fenarimol fungicide completely degraded in three weeks while all the other a.i. (active ingredent) showed considerable persistence. Except pyrazophos, an accumulation of residues left by repeated applications was observed when experiments were carried out on greenhouse tomatoes for studying the degradation of two insecticides, one acavicide, and four fungicides (Fenarimol pyrazophos and others).

Further more Cabras *et al*. (1985) reported that fenarimol residue became indetectable after three weeks while on tomatoes sprayed three time there was still a residue of about 0.1ppm

Also Carbras *et al*. (1985) found that pyrazophos showed a low level residue after single spraying but with considerable persistence after 21 days, only slight degradation occurred.

Metalaxy residues:

Data presented in table (2) showed that the collected samples from Bab El-Loak, Embaba, Dokki, Benha, Menouf, Dikerness, Zagazig, Fayed, and Minia regions were free from any residues of Metalaxyl. While those marketed from other regions recoded fungicide residue varied from 0.15ppm to 5.77ppm in Giza and Heliopiles, respectively.

Washing caused reduction of the obtained residues by 66.67% to 71.88% and gave higher decontamination of Metalaxyl residues.

This results is agreement with Rady *et al*. (1992) as they reported that washing cucumber fruits with tap water reduced Metalaxy residues by and 33.85 to 52.25

We deduced from previous, that unwashed and washed cucumber samples which marketed from eighteen different regions was lower than MRL (0.5mg/kg for Metalaxyl in cucumber fruits) except which marketed from Heliopilies and Rod El-Farag where residues were 1.66 and 0.77ppm, respectively.

Metalaxyl rapid Transolocation toward the upper parts of the plant of cucumber, as indicated by the high concentration found in the leaves after planting and the metalaxyl residues transport to fruits. This results are inagreement with those Marucchini *et al*. (1983) as they reported Metalaxyl was still present in the leaves of sunflower plants 90days after planting.

These results are inagreement with these of Harlan and Richard (1977) as found that samples were analyzed for possible ETU residue. None i.e. less than 0.05ppm was found when cucumbers samples for residue

studied were collected from 17 different locations throughout the united states where Ethylenethiourea (ETU) had been used as fungicides on cucumber fruits.

The use of these fungicides is increasing because their correct application dose not produce a residue content higher than the legal limits for harvested cucumber. However although a loss of fungicides occurs during washing process it is possible to find consistent residues of the fungicides in cucumber as a consequence of the frequent application of heavy dosages, together with or because of lack of respect for the statutory safety interval between treatment and harvest. This is agreement with (Cabras et al 1984 and Holland *et al.,* 1994)

In the greenhouse the fungicides active ingradients were stable, while in the field it was degraded moderately this results are inagreement with Cabras *et al.* (1995).

In case of repeated fungicides treatments, the remarkable stability of fungicides in cucumber fruits could lead to active ingradient accumulation in the fruits, thus causing high residues at harvest time. These obtained results are inagreement with data reported in Cabras *et al*. (1995).to decrease fungicides residues in cucumber fruits must be stoppage the frequent spraying on the vegetables. These results are inagreement with Nagami, H. (1997).

Washing process by tap water reduced the fungicides residues, reduction in fungicides residues emphasized the importance of the washing process in minimizing contomination of the cucumber fruits with fungicides.

The above results are in line with those reported by El-Sheamy and Ramadan (1990). Washing the treated cucumber fruits with tap water reduced fungicide bupirimate fungicide residues by 5.97 to 36.84%.

In conclusion, it can be said that, although these treatment for cucumber fruits by frequent spraying gave good results in controlling infestation, the level of contamination still higher, therefore, the use of funigicides should be restricted from direct and frequent application, and the contaminated cucumber fruits with fungicides must not be harvested until they were found to be free from any detectable residues.

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تتبع مستوى تلوث ثمار خيار الصوب البلاستيكية بالمبيدات الفطرية فى الأسواق المصرية (2)

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ترش ثمار الخيار فى الصوب البلاستيكية بالعديد من المبيدات الفطرية الجهازية والغير جهازية وذلك لمكافحة الأمراض الفطرية التى تصيب ثمار الخيار ولذلك فى هذا البحث ثم شراء عينات من ثمار الخيار خلال شهر إبريل لعام 2000 من عدد أسواق 18 تمثل 12 محافظة مصرية وهى محافظة القاهرة (مصر الجديدة – باب اللوق – روض الفرج – الحلمية القديمة) ومحافظة الجيزة (الجيزة – إمبابة – الدقى) ومحافظة الأسكندرية (العجمى) ومحافظة القليوبية (بنها) ومحافظة المنوفية (منوف) ومحافظة الدقهلية (دكرنس) ومحافظة الشرقية (الزقازيق) ومحافظة البحيرة (مديرية التحرير – كوم حماده) ومحافظة الأسماعيلية (فايد) ومحافظة بورسعيد (حى العرب) ومحافظة المنيا (ملوى) ومحافظة قنا (قنا) وتم تقدير متبقيات خمسة مبيدات فطرية هى : دانيكونازول ، كاربوكسين ، بيرازوفوس ، فنياريمول ، ميتالاكسيل على وفى ثمار الخيار الغير مغسولة والمغسولة بماء الصنبور جيداً وكانت النتائج المتحصل عليها كما يلى :-

1- إن ثمار الخيار الغير مغسولة والمغسولة بماء الصنبور والتى تم شرائها من 18 منطقة ومركز كانت خالية تماماً أو أقل من الحد المسموح به عالمياً من متبقيات مبيدات دانيكونازول وكاربوكسين بإستثناء التى تم شرائها من الحلمية القديمة ، مديرية التحرير حيث كانت كمية المتبقى من هذين المبيدين أعلى من الحد المسموح به عالمياً (0.5 جزء فى المليون).
2- ثمار الخيار الغير مغسولة والمغسولة بماء الصنبور والتى تم شرائها من 18 منطقة ومركز جميعها كانت خالية تماماً أو أقل من الحد المسموح به عالمياً من متبقيات مبيد ميتالاكسيل بإستثناء العينات التى تم شرائها من مصر الجديدة وروض الفرج حيث كانت كمية المتبقى من هذا المبيد أعلى من الحد المسموح به عالمياً (0.5 جزء فى المليون).
3- ثمار الخيار الغير مغسولة والمغسولة بماء الصنبور التى تم شرائها من 18 منطقة ومركز جميعها كانت خالية تماماً أو أقل من الحد المسموح به عالمياً من متبقيات مبيد فيناريمول بإستثناء العينات التى تم شرائها من العجمى ، بنها ، دكرنس حيث كانت كمية المتبقى من هذا المبيد أعلى من الحد المسموح به عالمياً (0.5 جزء فى المليون).
4- ثمار الخيار الغير مغسولة والمغسولة بماء الصنبور والتى تم شرائها من 18 منطقة ومركز كانت كمية المتبقى من مبيد بيرازوفوس أعلى من الحد المسموح بـــه عالمياً (0.1 جزء فى المليون) بإستثناء العينات التى تم شرائها من الحلمية القديمة ، إمبابة ، الدقى ، دكرنس ، الزقازيق ، كوم حماده ، حى العرب ، قنا فكانت خالية تماماً أو أقل من الحد المسموح به عالمياً من متبقيات هذا المبيد.

وعلى ذلك فأن ثمار الخيار التى تم شرائها من 18 منطقة ومركز من الأسواق المصرية غير صالحة للأستهلاك الأدمى بالرغم من غسلها جيداً بماء الصنبور بإستثناء ثمار الخيار التى تم شرائها من مناطق إمبابة ، الدقى ، الزقازيق ، كوم حماد ، حى العرب ، قنا حيث كانت خالية تماماً أو أقل من الحد المسموح به دولياً من متبقيات المبيدات الفطرية والتى تم تقدير متبقياتها وهى بذلك صالحة للأستعمال الأدمى . وبالتالى وجدت بعض ثمار الخيار خالية من المبيدات فى بعض الأسواق بينما وجدت أخرى ملوثة ببعض المبيدات التى قد تشكل خطورة للأنسان أو الحيوان نتيجة استهلاكها حيث أحتمال خطورتها من نواتج تكسير المبيد أو من المبيد الأصلى . حيث مقدار ما تحتفظ به ثمار الخيار من متبقيات المبيدات الفطرية عند رشها مرة واحدة أقل بكثير عنه فى حالة زيادة عدد مرات الرش فى الصوب البلاستيكية .ولذلك ينصح بعدم تكرار الرش بالمبيدات الفطرية على ثمار الخيار وبالذات بالفترات التى تسبق عملية الحصاد أو جمع المحصول تحت ظروف الصوب البلاستيكية .

Table (2): Level of Pyrazophos, Fenarimol, and Metalaxyl residues on and in unwashed and washed cucumber fruits (ppm)

Fungicides	Pyrazophos			Fenarimol			Metalaxyl		
Samples / Location	Unwashed	Washed	% Loss by washing	Unwashed	Washed	% Loss by washing	Unwashed	Washed	% Loss by washing
Heliopiles	0.95	0.42	55.79	0.39	0.05	87.18	5.77	1.66	71.23
Bab El-Loak	5.20	2.30	55.77	UND	UND	-	UND	UND	-
Rod El- Farag	0.53	0.24	54.72	0.05	0.02	60.00	2.68	0.77	71.27
El-Helmia El-Kadema	UND	UND	-	0.86	0.11	87.21	1.70	0.49	71.18
Giza	0.34	0.15	55.58	1.55	0.27	82.58	0.15	0.05	66.67
Embaba	0.13	0.05	61.54	UND	UND	-	UND	UND	-
Dokki	UND	UND	-	0.11	0.03	72.73	UND	UND	-
El-Agamy	1.11	0.45	59.46	4.20	0.73	82.62	1.12	0.33	70.54
Benha	3.04	1.47	51.64	4.45	0.57	87.19	UND	UND	-
Menouf	2.87	1.11	61.32	0.38	0.05	86.84	UND	UND	-
Dikerness	UND	UND	-	5.80	2.15	62.93	UND	UND	-
Zagazig	UND	UND	-	UND	UND	-	UND	UND	-
Modriet El-Tahrir	0.42	0.19	54.76	UND	UND	-	0.58	0.18	68.97
Kom Hamada	UND	UND	-	UND	UND	-	0.82	0.27	67.07
Fayed	0.50	0.20	60.00	0.38	0.07	81.58	UND	UND	-
El-Arab	UND	UND	-	UND	UND	-	0.32	0.09	71.88
Minia	0.85	0.39	54.12	UND	UND	-	UND	UND	-
Kena	UND	UND	-	UND	UND	-	1.45	0.48	66.90

UND = Undetectable