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Maximum Residue Limit and Risk Assessment of Spiromesifen (BAY BSN 2060; Oberon 240SC) on Tea (*Camellia sinensis* (L) O' Kuntze)

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Crop protection chemicals are highly regulated, their testing and regulation being equivalent to that of pharmaceuticals. Judicious uses of such chemicals maximize the farm, orchard productivity. These have been the most powerful tools available to mankind for combating pests of agriculture/ horticulture and public health. Simplicity, efficiency and economic return have been the hallmark of their wide use. As a result of continuous use, at times their residues find the place in soils, water and other environment and finally contaminate our food (Atreya 2001). Today's consumer expects an affordable constant yr round supply of clean, fresh, healthy and above all safe food and beverages.

In order to protect the health of the consumer while facilitating international trade, the primary objective is to develop the Maximum Residue Limit (MRL). Public health considerations are taken into account by establishing the MRL's at level not higher than those resulting from the use of pesticide in accordance with Good Agricultural Practices (GAP). However, explicit consideration of possible exposure to residues of pesticides is an integral part of the risk assessment process to ensure that Acceptable Daily Intake (ADI) of the pesticide is not exceeded. The best assurance that exposure to residues are within safe limits is obtained by dietary intake studies but when such studies is not feasible or the pesticide is not being used or has only recently been approved for use, pesticide residues intake must be predicted on the basis of available data (Anonymous 1989; 1997).

In India, Spiromesifen (BAY BSN 2060) is recently introduced and is presently under consideration of approval/ registration for use on plantation crops including tea. The MRL's for this insecticide (acaricide) on tea have not been fixed in our country. Therefore, an attempt has been made to fix its MRL on tea (made) and to study risk assessment by generating food chemical concentrations (or residues) data through supervised fixed trials conducted for successively three crop yr to determine i) rate of decline of spiromesifen deposits ii) highest levels of residues at harvest to estimate the potential exposure from proposed use of spiromesifen on tea bushes and iii) to predict its total dietary intake, consumer risk assessment and to propose MRL.

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MATERIALS AND METHODS

Tea (*Camellia sinensis* (L) **O Kuntze**) bushes (variety kangra local) in plots (5X5 m^2) at Tea Experimental Farm, Department of Tea Husbandry and Technology, CSK Himachal Pradesh Krishi Vishvavidyalaya Palampur (Himachal Pradesh), India were selected for the study. Plant spacing was 150X150 cm^2. Spiromesifen is recommended especially for mite pests like purple mite (*Calacarus carinatus* **Green**), red spider mite (*Oligonychus cofae* (**Niether**)) and also against hemipteran and homopteran pests (Liu 2004) at variable dosages from, 100 to 150 g ai ha^{-1}. In other countries also this has been found effective at varying dosages 100-150 g ai ha^{-1} as foliar spray (Nauen et al. 2002; 2003). In view of this spiromesifen (Oberon 240SC) (BAY BSN 2060, acquired from Bayer Crop Science Limited, New Delhi, India) was sprayed on tea bushes @ 112.5 g ai ha^{-1}, 150 g ai ha^{-1} (recommended dose) and 300 g ai ha^{-1}(double the recommended dose) during three yr trials (2002-2004). The spiromesifen (Oberon 240 SC) formulation was dissolved in water. The appropriate solution was applied using a hand operated knapsack sprayer with a spray volume of 750 L ha^{-1}. The experiment was laid in randomized block design and each treatment was replicated three times.

For determining spiromesifen concentration in green tea leaves and made tea (black and green) at different intervals, the two leaves and a bud samples (750 g) were harvested and collected randomly from each treatment on 0 d (1hr after pesticide spray), 1, 3, 5, 7, 10, 15, 21, 28 d from the spray. Harvested green tea leaves were divided in to three equal parts (one for extracting residues from green leaves, other two were used for manufacturing black and green tea).

Black tea was prepared from plucked green tea leaves. Green tea leaves were spread on Hessian cloth mounted on wooden frame at a thickness 2.5 cm and allowed to wither under ambient conditions for the period of 20 hr. The withered leaves were passed into a rolling machine (Pizy roller consisting of five miniature hoods, capacity 6 g each) for 1 hr. The rolled tea was spread over fermentation trays at a thickness of 1.3-1.8 cm for a period of 1 hr at 92$\pm$ 2 percent relative humidity. Fermented leaves were kept in drying hot air chamber at 90-95^{0}C for first 20-30 min and at 75-80^{0}C for next 25-30 min.

The green tea was manufactured by initially sterilizing leaves by passing steam through leaves for about 2 min. The leaves were cooled and dried in hot air at 90-100^{0}C for 4-5 min. The dried leaves were rolled initially for 15 min without heat followed by rolling at 50-60^{0}C for 30-40 min and 80-90^{0}C for 40 min. The rolled tea kept in hot air oven at 80^{0}C for 1 hr.

The tea leaves were cut into small pieces (approximately 1x1 cm^2), mixed thoroughly and a sub sample of 25 g was taken for the further processing. Soil samples (0-10 and 10-20 cm deep cores, 500 g) were collected after 7 d of spray

to examine the movement/ leaching in soil. Soil samples were air dried, ground and sieved and a sub sample of 25 g was taken for analysis.

Tea samples were extracted and cleaned up (Anonymous 2002). Recovery experiments utilizing all the substrates were carried out, in triplicates, at various fortification levels, by adding known volumes of spiromesifen (Oberon 98.9% purity acquired from Bayer Crop Science Limited, New Delhi, India). The recoveries of spiromesifen from spiked green tea leaves, made tea black, green were 97.4, 89.2, 89.4 percent, respectively; 87 percent from soil. Quantification of residues was carried out through gas liquid chromatography (PERKIN ELMER AUTO GC equipped with AUTOSAMPLER and PE 1022 INTEGRATOR, CAPILLARY COLUMN BP 225; Detector ECD; Temp. Oven 250^0C, Injector 250^0C, Detector 350^0C; Gas Flow N_2 40 m L min^{-1}, H_2 4 m L min^{-1}, Air 450 m L min^{-1}), RT 1.867 min. GLC has 0.001 mg kg^{-1} LOD (limit of detection) for spiromesifen.

Since MRL of spiromesifen are not available on tea in India, an effort was made to establish safety factors like Pre- harvest Intervals (PHI), Theoretical Maximum Residue Contribution (TMRC) and Predicting Dietary Intake (PDI) in order to arrive at risk assessment of human health from analytical data as described by Borah et al. (2003); Dikshit et al. (2003).

RESULTS AND DISCUSSION

The data on the availability of spiromesifen in/on green tea leaves and made tea (black and green) presented in Table 1, 2, 3 were viewed for different guiding parameters to find the safety assurance and risk assessment *vis-a- vis* treatments by comparing the dietary intake exposure, which can be reduced to low and occasional (Bates 2002). The actual exposure of any consumer to pesticide residues can theoretically be determined by the analysis of the consumer's total diet.

The initial deposits of 6.22-7.04, 10.36-11.71 and 19.99-23.01 mg kg^{-1}, respectively of spiromesifen in/on green tea leaves declined progressively with time from all the treatments/ trials and became non detectable on 21 d of application at lower doses (112.5 and 150 g ai ha^{-1}) and 28 d of application at higher dose (300 g ai ha^{-1}). The initial deposits in made tea was low due to major loss during the process of manufacturing from green tea leaves in to made tea.

The ADI (Acceptable Daily Intake) of Oberon is 0.022 mg kg^{-1}body weight. Therefore MPI (Maximum Permissible Intake) i.e. maximum permissible quantity of insecticide for human being (50 kg, international low weight consideration) per day during a part or whole life is 1.1 mg $person^{-1}day^{-1}$; for a person of 55 kg weight, the MPI is 1.21 mg $person^{-1}day^{-1}$ without involving any appreciable risk or at no-observed-adverse-effect-level (NOAEL). Considering the average tea consumption of processed tea by a person in a day to be 25 g i.e. 0.025 kg out of a

total of 1.5 kg food consumed per day per person, the TMRC (Theoretical Maximum Residue Contribution)/ TMDI (Theoretical Maximum Daily Intake) value from green tea leaves collected on 3 d from recommended dose i.e. 150 g ai ha^{-1} found to be 0.099 mg $person^{-1}day^{-1}$(based on 3 yr trials supervised median pesticide residues). The calculated TMDI value is much lower than the MPI value, therefore spiromesifen treatment of recommended dose appears safe and consumption of such tea is not likely to risk the health. Even if higher treatment rate (300 g ai ha^{-1}) and the corresponding median pesticide residue value is considered the TMDI value (0.158 mg $person^{-1}day^{-1}$) is found lower than the MPI value, therefore higher dose, if used inadvertently would also not probably cause harm to consumers. But still to have more Margin of Safety (MOS) and as a normal convention in India, the tea leaves are not plucked just after the spray.

Table 1. Availability of spiromesifen in/on unprocessed green tea leaf samples after foliar application.

Days	Treatments (g ai ha^{-1})	Amount of spiromesifen (mg kg^{-1})		
		2002	2003	2004
0	112.5	6.22	7.04	6.34
	150	10.36	11.71	10.42
	300	21.36	23.01	19.99
1	112.5	4.75 (23.69)	6.756 (4.10)	5.05 (20.43)
	150	8.17 (21.13)	10.84 (7.44)	7.84 (24.75)
	300	15.39 (27.92)	19.15 (16.74)	13.11 (34.40)
3	112.5	2.57 (58.59)	2.69 (61.70)	3.14 (50.49)
	150	5.27 (49.13)	5.99 (48.80)	4.66 (55.25)
	300	9.20 (56.92)	9.51 (58.66)	8.08 (59.55)
5	112.5	1.76 (71.63)	1.87 (73.34)	1.22 (80.74)
	150	3.26 (68.45)	3.33 (71.56)	3.71 (64.38)
	300	5.11 (76.06)	5.52 (75.99)	5.35 (73.22)
7	112.5	1.05 (83.07)	1.33 (81.06)	1.04 (83.59)
	150	2.27 (78.05)	2.61 (77.72)	2.24 (78.45)
	300	3.54 (83.41)	2.93 (87.25)	3.29 (83.50)
10	112.5	0.81 (86.94)	0.83 (88.11)	0.90 (85.77)
	150	1.17 (88.62)	1.59 (86.42)	1.22 (88.21)
	300	2.17 (89.80)	1.66 (92.78)	2.48 (87.57)
15	112.5	0.21 (96.57)	0.27 (96.09)	0.24 (96.09)
	150	0.54 (94.33)	0.57 (95.05)	0.41 (96.02)
	300	1.22 (94.28)	0.84 (96.32)	1.41 (92.94)
21	112.5	ND (100)	ND(100)	ND(100)
	150	ND (100)	ND(100)	ND(100)
	300	0.30 (98.57)	0.11	0.11 (99.40)
28	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	ND(100)
	300	ND(100)	ND(100)	ND(100)

ND, Non detectable; Figures in parentheses are percent cumulative loss.

Table 2. Availability of spiromesifen in/on mad tea black after foliar application.

Days	Treatments (g ai ha^{-1})	Amount of spiromesifen ($mgkg^{-1}$)		
		2002	2003	2004
0	112.5	5.28	6.97	5.54
	150	7.89	10.74	8.05
	300	14.04	21.93	14.10
1	112.5	4.54 (14.01)	6.34 (9.02)	3.49 (36.94)
	150	6.74 (14.47)	9.70 (9.68)	5.56 (30.94)
	300	12.66 (9.83)	18.43 (15.96)	11.70 (17.02)
3	112.5	2.80 (46.96)	2.20 (68.35)	2.15 (61.20)
	150	4.13 (47.56)	5.83 (45.72)	4.00 (50.39)
	300	7.43 (47.09)	8.43 (61.55)	7.05 (50.02)
5	112.5	2.54 (51.79)	1.54 (77.90)	0.77 (86.05)
	150	3.24 (58.85)	3.16 (70.59)	2.99 (62.76)
	300	5.05 (64.02)	5.46 (75.07)	4.46 (68.33)
7	112.5	1.34 (74.45)	0.96 (86.22)	0.50 (90.89)
	150	1.98 (81.21)	1.57 (85.39)	1.93 (75.97)
	300	3.91 (72.11)	3.30 (84.94)	2.19 (84.43)
10	112.5	0.23 (95.51)	0.16 (97.63)	0.06 (98.91)
	150	0.48 (93.87)	0.79 (92.64)	0.59 (92.63)
	300	0.91 (93.47)	1.25 (94.30)	1.58 (88.80)
15	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	0.004 (99.96)	0.28 (96.52)
	300	0.08 (99.42)	0.01 (99.92)	0.82 (94.17)
21	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	ND(100)
	300	ND(100)	ND(100)	ND(100)
28	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	ND(100)
	300	ND(100)	ND(100)	ND(100)

ND, Non detectable; Figures in parentheses are percent cumulative loss.

The tea leaves (two leaves and a bud) are normally harvested after 7 d of application of spray (minimum 5-7 d after spray). In view of this, the TMRC/ TMDI values from the recommended treatment based on STMPR (Supervised trial median pesticide residues) from the 3 yr data, on 5 d, found to be 0.061 mg $person^{-1}day^{-1}$. This value is still much lower than the MPI value and also lower than the TMRC/ TMDI from green leaves after 3 d of spray. Therefore application of spiromesifen appears safe on this crop and probably involves no risk to consumers and free from health hazards. A minimum of 3 d pre harvest interval is suggested and on the basis of extrapolation as above and the facts known at that time MRL of 6.62 mg kg^{-1} for tea.

Half life (Table 4) of spiromesifen ranged from 2.86-3.62, 1.42- 3.54, 1.73-3.46 d respectively in unprocessed green tea leaves, made tea black and made tea green. Bioefficacy studies revealed that spiromesifen treatments (112.5 and 150 g ai ha^{-1})

Table3. Availability of spiromesifen in/on made tea green after foliar application.

Days	Treatments (g ai ha^{-1})	Amount of spiromesifen ($mgkg^{-1}$)		
		2002	2003	2004
0	112.5	5.14	6.68	5.40
	150	7.55	9.97	7.87
	300	12.04	21.37	13.98
1	112.5	4.33 (15.75)	5.99 (11.44)	4.10 (24.01)
	150	5.67 (27.87)	9.24 (7.30)	5.64 (28.23)
	300	10.66 (11.41)	19.41 (10.68)	10.77 (22.92)
3	112.5	2.66 (48.32)	2.50 (62.49)	2.09 (61.27)
	150	3.91 (48.18)	4.29 (56.90)	3.98 (49.36)
	300	7.89 (34.46)	9.33 (57.06)	7.64 (45.31)
5	112.5	2.29 (55.35)	1.86 (72.18)	0.81 (84.99)
	150	2.85 (62.23)	3.09 (69.00)	2.68 (65.90)
	300	4.85 (59.71)	5.18 (76.16)	4.86 (65.20)
7	112.5	1.12 (78.07)	1.19 (82.20)	0.59 (88.95)
	150	1.45 (80.75)	2.23 (77.64)	1.99 (74.60)
	300	3.91 (67.48)	2.68 (87.66)	2.13 (84.75)
10	112.5	0.16 (96.75)	0.72 (89.23)	0.05 (99.01)
	150	0.38 (94.90)	1.47 (85.21)	0.48 (93.81)
	300	0.88 (92.698)	1.23 (94.34)	1.24 (91.07)
15	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	0.23 (95.21)
	300	0.06 (99.49)	0.35 (98.38)	0.40 (97.08)
21	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	ND(100)
	300	ND(100)	ND(100)	ND(100)
28	112.5	ND(100)	ND(100)	ND(100)
	150	ND(100)	ND(100)	ND(100)
	300	ND(100)	ND(100)	ND(100)

ND, Non detectable; Figures in parentheses are percent cumulative loss.

Table 4. Half life (days) of spiromesifen in/on tea.

Type of tea	Treatments (g ai ha^{-1})	Half life (days)		
		2002	2003	2004
Un processed tea	112.5	3.23	3.34	3.34
	150	3.50	3.42	3.30
	300	3.62	2.86	3.20
Made tea (black)	112.5	2.42	1.91	1.60
	150	2.64	1.42	3.04
	300	2.06	1.55	3.54
Made tea (green)	112.5	2.18	3.01	1.73
	150	2.46	3.46	1.86
	300	2.02	2.44	2.89

were more effective (reduces mite population >90 percent over untreated check) in checking the menace of both the species of mites (*C. carinatus* and *O. cofae*) on tea prevalent in Himachal Pradesh for 3-4 weeks of spray compared to dicofol 185 g ai ha^{-1} (reduces mite population only 69- 80 percent). Spiromesifen persisted for up to 7 d at levels found effective in checking mite population. Soil samples (0-20 cm), collected at final harvest of crop, did not show the presence of insecticide residues and therefore no build up and downward movement is expected.

The double the recommended dose did not exhibit any phytotoxic effect during the growth of the crop. The intended doses (recommended, 150 gai $ha^{-1}$and double the recommended, 300 g ai ha^{-1}) did not cause any appreciable effect on beneficial insects like *Coccinella* sp. and *Syrphis* sp.

The pesticide spiromesifen was effective in managing pests and found to be short lived on tea. Based on environmental evaluations, consumer health risks are minimal at the recommended doses.

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