International Journal of Chemical Studies

P-ISSN: 2349–8528
E-ISSN: 2321–4902
IJCS 2019; SP6: 698-702

Ranjit Singh
Indian Institute of Natural Resins and Gums, Ranchi-Jharkhand, India

Anandakumar S
Indian Institute of Crop Processing Technology, Thanjavur, India

Corresponding Author:
Ranjit Singh
Indian Institute of Natural Resins and Gums, Ranchi-Jharkhand, India

(Special Issue -6)
3rd National Conference
On
PROMOTING & REINVIGORATING AGRI-HORTI, TECHNOLOGICAL INNOVATIONS [PRAGATI-2019]
(14-15 December, 2019)

Effect of ozone fumigation on controlling drugstore beetle and quality of coriander during storage

Ranjit Singh and Ananda Kumar S

Abstract
Coriander (*Coriandrum sativum*) the spice comes under minor aromatic annual condiment spice crop. Coriander leaves and fruits are used in flavouring food and to suppress offensive odours in pharmaceutical preparations. Several insect damage dried coriander fruits during storage. Among the insects, cigarette beetle (*Stegobium paniceum*) is important one. Now a day, ozone fumigation is gaining interest because it can be generated on the treatment site also not produce any residue after the treatment. In the study, four life stages of drug store beetle such as adult, pupa, larva and egg were fumigated in lab-scale canisters by ozone at different concentrations like 150, 250 and 350ppm to achieve 100 per cent mortality. From the results, it is observed that the adult insect has taken 578min to attain LT99 at 250 ppm. Similarly, for pupa and larva the 99 per cent mortality was achieved at 1083min and 609min respectively. The ozone treated (250ppm) coriander samples were kept in airtight plastic containers and storage studies were conducted for 105 days. During the storage, it was observed that there was significant decrease in the quality parameters of protein, colour b value and linalool content. The respective quality value of 250ppm treated coriander on the 105th day of storage were found as protein 12.25 per cent; colour (b value) 23.71 and linalool content reduced to 37.48 per cent.

Keywords: Ozone fumigation, Coriander, Drugstore beetle, Probit analysis, Quality, Protein, and linalool content

1. Introduction
spices and condiments have always been considered a legacy of India and India still continues to be largest producer, consumer and exporter of spices in the world. Coriander (*Coriandrum sativum*) the spice comes under minor aromatic annual condiment spice crop. The major varieties cultivated in India are Gujarat coriander-1, Gujarat coriander-2, RCr-41, UD-20, RAJENDRA swati (RD-44), CS-287, CO-1, CO-2, CO-3, Sadhana, Swati and Sindhu (Spice Board of India, 2012-2013). Today, most coriander is consumed in the form of curry powder (Purseglove *et al*., 1981).In India, coriander seeds are extensively employed as a condiment in the preparation of pickling spices, sausages and seasonings, and for flavouring pastry, cookies, buns and cakes, and tobacco products. The factors which affect the quality of coriander during storage are poor maintenance and lack of sanitation in storage facilities. Storing the coriander for longer time is unfeasible, because the quality of the coriander is deteriorated by the attack of pests which includes birds, rodents, insects and microorganisms (Rao *et al*., 1975) [15]. *Stegobium Paniceum* (l.) commonly known as drugstore beetle (known in the United Kingdom as the biscuit beetle). As name indicates, it's a pest for both herbal medicines and stored biscuits.

Adults *S. Paniceum* (l.) are cylindrical, 2.25 to 3.5 mm (1/10 to 1/7 inch) long, and are a uniform brown to reddish brown. They have longitudinal rows of fine hairs on the elytra. Van Emden (1929) [18] considered that *S. Paniceum* could live on material whose moisture content is between 6 and 15 per cent. Development of the beetles is possible at temperatures between 15 and 35 °C and 30% RH and above. Optimum conditions for rapid development are 30 °C and 60 to 90% RH (Lefkovitch, 1967) [11]. The insect has been known to attack grain and grain products, spices and herbs, dried fruit, seeds, dried fish, bread, birdseed, dry dog and cat food, coffee beans, chocolate, powdered milk, and many other organic materials (Hagstrum and Subramanyam, 2009) [6].

Fumigation of godowns with phosphine gas from celphos tablets at a rate of 140 tablets/100 m^3 resulted in mortality of adults and larvae (Kavadia *et al.*, 1978) [10]. Jacob (1986) [9] suggested that infestation of pest can be controlled when Aluminium Phosphide is used as a fumigant in an airtight store for 2 to 3 days. Now a day, ozone finds wide application as a powerful oxidant and it has numerous beneficial applications in the food industry. Ozone, a known sterilant, can be used as an insect control agent in food commodities. Ozone is readily generated from atmospheric oxygen and is safe to the environment when used for fumigation. However it is highly unstable and breaks down to molecular oxygen quickly (Mason *et al*., 1999; Hollingsworth, and Armstrong, 2005) [12, 8]. Hence, the study was conducted on Effect of ozone in controlling *Stegobium Paniceum* (l.) and quality of Coriander seeds during storage.

2. Materials and Methods

Coriander seeds (Variety: UD – 21) were procured in bulk quantity from the local market in Thanjavur district of Tamil Nadu, India. The initial moisture content of the coriander was determined using hot air oven method (AOAC, 2000) [3].

2.1 Ozone Fumigation System

The ozone fumigation system (Fig. 2.1) consists of oxygen concentrator, ozone generator, fumigation chamber (canister), ozone analyzer and ozone destructor. The lab scale canisters were developed using PVC pipe to carry out the ozone fumigation study. The dimension (height × diameter × thickness) of canister is 390mm ×60mm m × 2mm. Both the ends were tightly closed with the help of end cap and the ends were sealed in order to avoid leakage of ozone gas during fumigation. The ozone inlet and outlet of pipes were connected on bottom and top end of canister respectively. The diameter of inlet and outlet pipe was 7mm. For uniform distribution of ozone gas inside the canister and for placing of samples in a canister a perforated sample holder was designed with the help of SS wire mesh, supported by metallic rods.

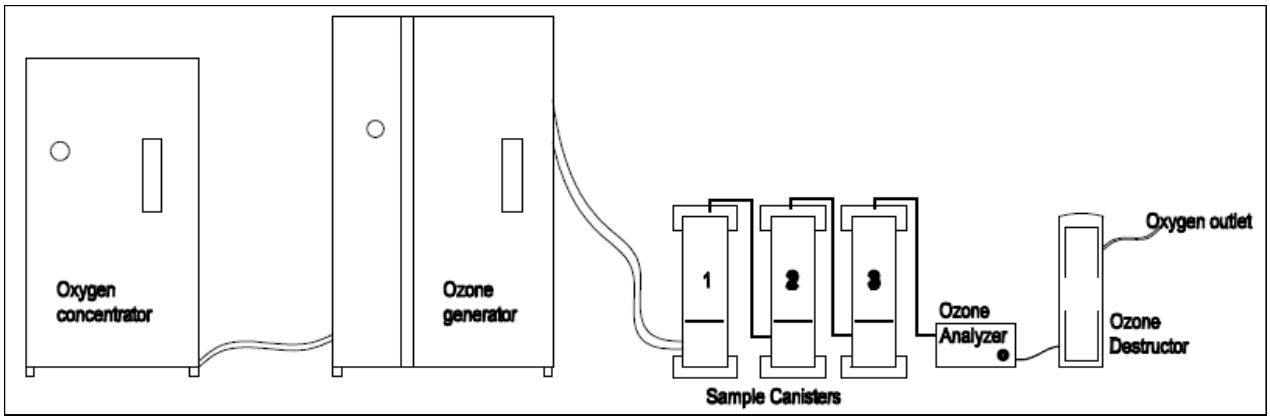

Fig 2.1: Schematic diagram of Ozone Fumigation System

100g of coriander seeds were placed into the canisters. The generated ozone gas from ozone generator was passed into canister through the inlet. The incoming ozone gas concentration was measured with ozone analyzer. The canister outlet was connected to the ozone destructor, where the ozone gas gets destructed into oxygen before deliver to the environment. The three canisters are connected in series for each experiment with three replications for at same concentration study.

2.2 Test insect

Stegobium paniceum (L.). was provided by Department of Primary Processing, storage and Handling, Indian Institute of Crop Processing Technology. Insects were incubated at 30±2 °C, 75±5% R.H and reared on whole wheat flour containing 5% yeast.

2.3. Toxicity of ozone at different stages of *Stegobium paniceum* (L.).

Ten numbers of adult, pupae, larva and egg of *Stegobium paniceum* (L.), and 100 g of coriander seeds were placed into the each canister. The ozone gas was allowed to pass through the canister with different concentration and different exposure time until it attains 100% mortality. The experiment was performed with three replicates for each treatment at 10 per cent moisture contents of coriander. The treated adult, pupae, larva were placed in wide opened glass jar with 10g of feed and kept in environmental chamber set at 30±2 °C, 75±5% RH and the mortality rate of insects were examined.

For larvae, the ozone treated larvae in the cages were incubated and daily observations were made until larvae were dead or turn to pupa. For pupae, the treated pupae in the cages were incubated and daily observations were made until pupae were dead or adult emergence. For adults, the treated cages were incubated for 24h to observe their survival under the stereomicroscope. The adults that did not show any response to touching using a brush were considered as dead. For every 30 min interval at different ozone concentration treatment the number of dead and live insects were recorded, and expressed in per cent mortality. The mortality rate of insects was calculated by using the formula (Omotoso, 2005) [13]

$$\text{Mortality (\%)} = \frac{\text{Number of Dead Insects}}{\text{Total Number of Insects}} \times 100$$

2.4 Quality Analysis

Biochemical analyses are important to assess the quality traits of the coriander and to evaluate their relative loss due to ozone treatment on storage periods. The various analysis *viz*., protein, colour b and linolool content of both control and ozone treated coriander was carried out by adopting standard procedures. The each experiment was carried out in triplicates and the average values were recorded.

2.4.1 Protein

The micro-Kjeldahl nitrogen method was used, which involved the digestion of 0.5g of sample with sulphuric acid and a catalyst followed by colorimetric determination of nitrogen. The value of nitrogen was multiplied by 6.25 to obtain percentage crude protein (AOAC, 1990) [2].

2.4.3 Colour 'b' value

Hunter lab Color Flex EZ meter (Hunter Associates Laboratory, Inc., Reston, Virgina, USA) was used for the measurement of colour value of coriander. Chromatic portion of the solids is defined by a (+) redness, a (-) greenness, b (+) yellowness and b (-) blueness. The colour of control and ozone treated coriander was measured at different storage periods at 10° observer at D65 illuminant ASTM E 308. The colour of the sample was measured by filling the coriander seeds in the transparent quartz cup without any void space at the bottom. All the treatments were replicated thrice and the average value was taken for further studies.

2.4.4 Linalool content

Linalool (3,7-dimethyl-1,6-octadien-3-ol) is a monoterpene alcohol, which occurs naturally in several aromatic plants (Shang *et al.*, 2002) [16]. Linalool is a much sought-after compound in the flavor and fragrance industry, and contributes to the characteristic aroma of a vast number of natural products, such as fruits and spices, as well as tea and chocolate. Linalool in plant samples was usually determined by GC-MS (Wang *et al.*, 2008) [19]. Twenty five, gm of coriander sample was soaked with overnight in ethanol. The concentration of 25 gm was selected based on availability of target compound peak in GCMS. GC-MS analysis of this extract was performed using a Perkin Elmer GC Claurus 500 system which is interfaced to a Mass Spectrometer equipped with a Elite-5 MS fused silica capillary column (30m x 0.25mm x 0.25µm df) composed of 5 percent Diphenyl and 95 per cent Dimethyl poly siloxane. Mass spectra were taken at 70eV; a scan interval of 0.5 seconds and fragments from 45 to 450Da. The relative percentage amount of each component was calculated by comparing its average peak area to the total areas.

2.4.4 Statistical Analysis

PROC PROBIT method was used to compute the half lethal time LT50 and 99% lethal time LT99 at 95% fiducial limits. The experimental results were analyzed by the statistical methods using the AGRES software (7.01) (Pascal Intel software solutions, India).

3. Results and Discussion

The effect of ozone fumigation at different concentrations and exposure time on mortality of Stegobium *paniceum* was determined at 7 per cent moisture content of coriander. The results are discussed below

3.1 Effect of Ozone on Mortality of *Stegobium paniceum* (L.)

The mortality of *Stegobium paniceum* (L.) at different ozone fumigation level and exposure time with respect 7 per cent moisture content was studied. The 100 percent mortality of adult at 150, 250 and 350 were achieved at 1219-minute, 578 minute and 443 minute respectively. The probit analysis of adults was studied (Table 3.1). The highest fudicial limit for L50 and L99 was recorded for 7 percent moisture content at 150, 250 and 350 ppm were 524.47, 360.21 and 246.80; 1219.22, 577.70 and 442.67 min respectively. The mortality data of adults tested with ozone fumigation agreed with those of several authors who observed toxicity of ozone against different stored products insect pests (Erdman 1980; Hasan *et al*. 2012) [4, 7]. Similarly the 100 percent mortality of pupa at 150, 250 and 350 were achieved at 1113-minute, 1083 minute and 809 minute respectively. The 50 percent mortality was achieved until 657 min, 476 min and 386 minute respectively. Also the mortality rates of larva were increasing with concentration level. The time of exposure was decreases with increase ozone concentration. The 100 percent mortality of pupa at 150, 250 and 350 were achieved at 725-minute, 609 minute and 561 minute respectively. The 50 percent mortality was achieved until 336 min, 260 min and 230 minute for 150, 250 and 350 ppm respectively. Sousa *et al*. (2008) [17] observed similar result related to pupa mortality tested with ozone fumigation against different stored products insect pests. From the ANOVA, it is observed that the mortality of *Stegobium paniceum* (L.) has not significant effect at different moisture content for L50 and L99. Considering the ozone concentration it has significant ($P<0.05$) differences.

Table 3.1: Probit Analyses on Mortality of *Stegobium paniceum* (l.) in different stages (Adult, Pupae and Larva) in Coriander at 7 per cent Moisture Content

Stages	O3 Concentration (PPM)	LT50 (95% Fiducial limits) (min)	LT99 (95% Fiducial limits) (min)	Slope ± SE	$\chi 2$ (df)
Adult	150	524.47 (503.39 - 545.54)	1219.22 (1112.47 – 1366.76)	6.35 ± 0.38	15.15 (30)
	250	360.21 (342.62- 377.54)	577.70 (541.93– 624.68)	6.24 ± 0.42	13.88 (22)
	350	246.80 (225.36- 268.02)	442.678 (396.71– 513.19)	5.05 ± 0.38	23.23 (16)
Pupae	150	657.07 (634.44 – 679.70)	1112.86 (1032.56 – 1229.89)	6.96 ± 0.63	5.71(31)
	250	476.22 (451.63- 501.10)	1083.35 (960.73– 1275.68)	6.51 ± 0.54	8.44 (25)
	350	385.67 (364.85- 406.07)	809.17 (727.04– 935.65)	7.22 ± 0.61	4.608 (22)
Larvae	150	336.16 (316.21 – 355.58)	725.28 (644.99 – 853.415)	6.96 ± 0.63	3.63 (19)
	250	260.17 (241.66- 278.31)	609.53 (530.42– 743.41)	6.29 ± 0.62	4.16 (15)
	350	230.48 (209.98- 252.01)	560.99 (461.74– 772.44)	6.02 ± 0.70	11(16)

3.2 Quality parameters of Coriander during storage

The quality of coriander such as protein content, color b value and linalool content were evaluated for control and ozone treated coriander during the storage period. The results are discussed below

3.2.1 Effect on Protein

The protein content of ozone fumigated coriander seeds at different concentration is shown in Fig. 3.1 from the graph it is observed that protein content of coriander seeds is decreasing with increase in the storage period. Among the ozone concentration the lower concentration 150 ppm treated sample has retained protein compared with 250 and 350 ppm. The protein content of coriander seeds at initial stage was 12.52 per cent. The protein content of adult treated coriander seed of 105 days storage period were 12.23, 12.25 and 12.22 per cent at 150, 250 and 350 ppm respectively. The loss in the protein content of coriander seeds stored in open atmosphere is high compare to ozone treatment. The lowest protein was found as 11.74 percent at end of 105 storage days in open atmosphere. Also, it was found that there was significant ($P<0.05$) effect of all process parameter moisture content, ozone dose and insect stages on protein content. Anandakumar *et al*., (2016) [1] has reported similar results for ozone treatment of turmeric rhizome during storage.

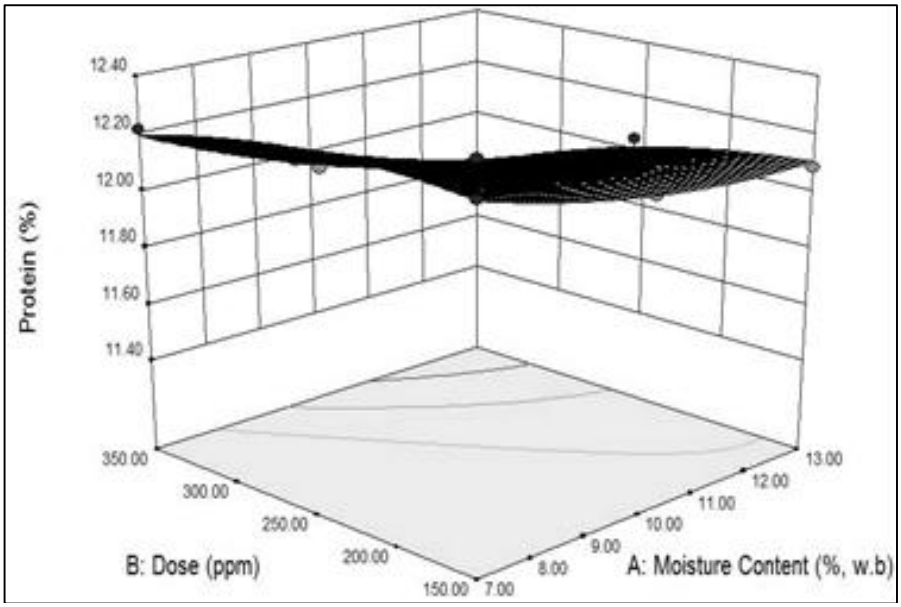

Fig 3.1: Effect of Ozone Fumigation on Protein Content Coriander

3.2.2 Effect on Colour b value

The colour value of ozone fumigated coriander seeds at different concentration is shown in Fig. 3.2. From the graph it is observed that b value of coriander seeds is decreasing with increase in the storage period. The b value of adult treated coriander seeds for 105 days storage period at 150, 250 and 350 ppm was 24.11, 23.71and 23.49 respectively. The initial b value was 25.22. The lowest b value was found as 24.11at end of 105 storage days in open atmosphere. Also, it was observed that there was significant ($P<0.05$) difference in the color b value during the storage. Similar results were reported by Gunasekaran (2001) [5] during Carbon dioxide fumigation of infested coriander powder, sambar powder and turmeric rhizomes storage studies.

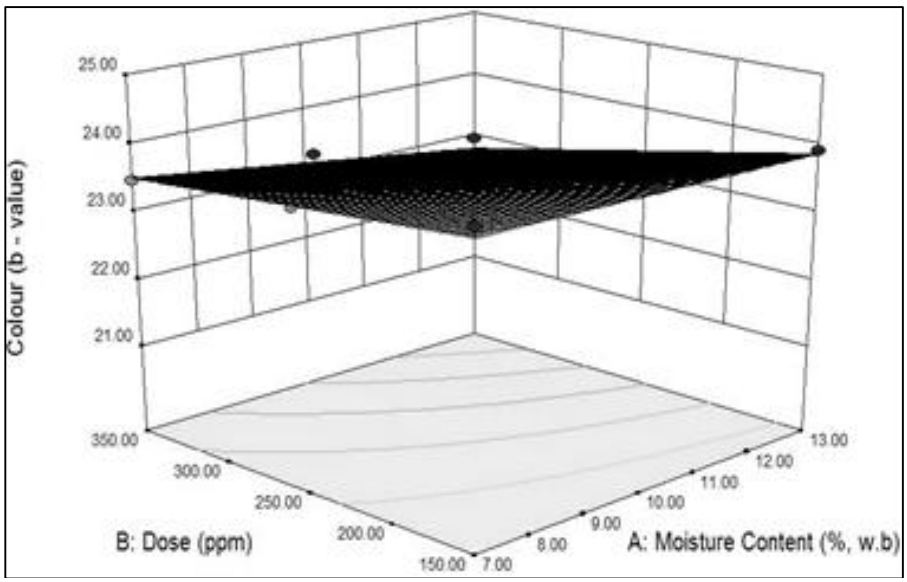

Fig. 3.2: Effect of Ozone Fumigation on Colour b value of Coriander

3.2.3 Effect on Linalool content

The effect on ozone fumigation in reduction of linalool component on 105 storage days is given in Fig. 3.3. It was observed that the linalool content was decreasing with increase in ozone concentration and time of exposure. The reduction in linalool content was found less in 150 ppm fumigated coriander for 10 hours and it was recorded the relative linalool content as 41.73 per cent.

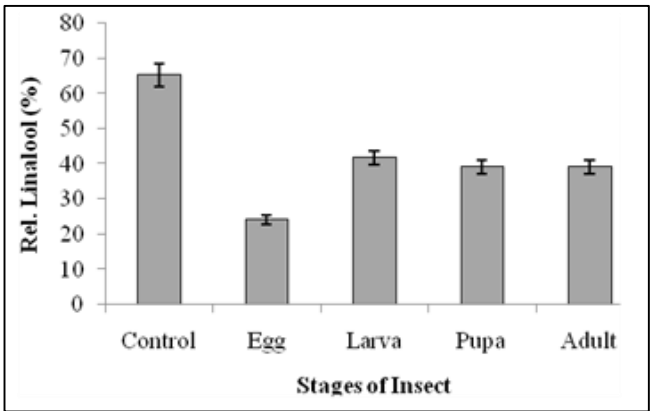

Fig 3.3: Effect of Ozone Fumigation on Reduction in Linalool content of Coriander at different Life Stages of *S. Paniceum (L)*

4. Conclusions

Three life stages of *Stegobiumpaniceum* (l.) were individually fumigated by ozone to achieve 100 per cent mortality. From the study it was observed that when the concentration of ozone increased, exposure time required to attain specific mortality was reduced. Among the treatments and stages the pupae stage has taken longer time to attain 100 per cent mortality. The quality of coriander like protein, Colour 'b' value and linalool was decreasing significantly ($P<0.05$) with increasing the ozone concentrations and storage period. The linalool content was found 41.73 per cent in 150ppm treated sample. These quality losses were found more in control samples than ozone treated samples during storage period.

5. References

1. Anandakumar S, Subhasini S, Alagusundaram K, Kavitha Abirami CV. Effect of Ozone fumigation on *Laiodermaserricorne* (F) and quality of turmeric rhizome, Journal of grain storage research. 2016, 126-132
2. AOAC Official Methods of Analysis of the Association of Official Analytical Chemists, 15th edition, Arlington, Virginia USA, 1990; 1, 2.
3. AOAC. Official methods of analysis of the Association of Official Analytical Chemists 16th edition, Arlington Virginia 22201. USA, 2000; 1, 2.
4. Erdman HE. Ozone toxicity during octonegy of two species of flour beetles, *Triboliumconfusum*and *T. castaneum*. Environmental Entomology. 1980; 9(1):16-17.
5. Gunasekaran N. Insect Control in Selected Spice Products Using Carbon Dioxide (Doctoral dissertation, University of Mysore), 2001.
6. Hagstrum D, Subramanyam B. Stored-product Insect Resource. AACC Press, St. Paul, 2009.
7. Hasan M, Phillips TW, Aikins MJ. Potential for ozone fumigation against anobiid beetles infesting stored products as an alternative to methyl bromide, Proceedings of 9th International Conference on Controlled Atmosphere and Fumigation in Stored Products, 15 - 19 October 2012, Antalya, Turkey. ARBER Professional Congress Services, Turkey, 2012, 260-265.
8. Hollingsworth RG, Armstrong JW. Potential of Temperature, Controlled Atmospheres and Ozone Fumigation to Control Thrips and Mealybugs on Ornamental Plants for Export. Journal of Economic Entomology, 2005; 98:289-298.
9. Jacob S. Host food preference of the cigarette beetle, *Lasiodermaserricorne* (F.) to few stored spices. *Plant Protection Bulletin*, 1993; 44:16-17.
10. Kavadia VS, Pareek BL, Sharma KP. Control of *Lasiodermaserricorne* Fab. Infestation of dry turmeric by phosphine fumigation. Entomology. 1978; 3:57-58.
11. Lefkovitch LP. A laboratory study of *Stegobiumpaniceum* (L.) (Coleoptera: Anobiidae). Journal of Stored Product Research. 1967; 3:235-249.
12. Mason LJ, Strait CA, Woloshuk CP, Maier DE. Controlling stored grain insects with ozone fumigation. In: Zuxun, J., Quan, L., Yongsheng, L., Xianchang, T., Lianghua, G. (Eds), Proceedings of the Seventh International Working Conference on Stored Product Protection, Beijing, China, October 1998. Sichuan Publishing House of Science and Technology, Chengdu, 1999, 536-547.
13. Omotoso OT. Insecticidal and insect productivity reduction capacities of aloe vera and Bryophyllum pinnatum on triboliumcastaneum (herbst). African Journal of Applied Zoology and Environmental Biology. 2005; 7(1):95-100.
14. Purseglove JW, Brown EG, Green CL, Robbins SRJ. Spices. Vol. 2. Longman, London, 1981; 736-788.
15. Rao RM, Reddy RCK, Subbarayadu M. Promising turmeric types of Andhra Pradesh, *Indian Spices*, 1975; 12:2-5.
16. Shang CQ, Hu YM, Deng CH, Hu KJ. Rapid determination of volatile constituents of *Michelia alba* flowers by gas chromatography–mass spectrometry with solid-phase microextraction. J Chromatogr. A, 2002; 942:283-288.
17. Sousa AH, Faroni LRA, Guedes RNC, Tótola MR, Urruchi WMI. Ozone as a management alternative against Phosphine-resistance insect pests of stored products. Journal of Stored Products Research. 2008; 44(4):379-385.
18. Van Emden F. Ueber die Rolle der Feuchtigkeit in Leben der Speicherschidlinge. Alrz. Schiidlingsk. 1929; 5:58-60.
19. Wang XY, Liu MC, Yang YW, Deng W, Li ZG. Chemical constituents of the essential oil of *Michelia alba* DC by GC-MS. J Chongqing Univ. 2008; 31:97-100.