Quality Characteristics of Various Propolis Sources from Commercial Products

王美月、徐泰浩 ; 張世揚

E-mail: 9115141@mail.dyu.edu.tw

ABSTRACT

Propolis is a natural resinous substance collected by bees from buds, young leaves and jnjurious parts of plants. The chemical composition and biological activity of compound of propolis varies greatly and directly on the local flora and phenology of the host plants and indirectly on the locality and season of collection. The objectives of this study were to analyze the chemical composition, chemical and to approve the antimicrobial ability of 9 crude propolis from different source and 79 commercial products. The chemical composition analysis includes determining of resin concentration, maximum UV spectrum, total flavonoids, total phenolic substances, heavy metal of lead , added tea, tartrazine, sunset yellow FCF of total flavonoids and antimicrobial ability for Staphylococcus aureus. Results show that the average resin of crude propolis was 29.50, from 16.21 to 53.81％, average resin of 79 commercial products was 28.58％, among average resin of 46 commercial ethanolic extract products containing propolis was 25.75 ％. Ultraviolet spectrum of crude propolis from different regions to maximum of wave length about 360~380 nm, was single peak, but Thailand region to wave length about 230 nm, was double peak. Commercial products to maximum of wave length have obvious difference. The total flavonoids average concentration of crude propolis was 17.91, the total flavonoids average concentration of commerical products was 9.03％. The total flavonoids of acced tea , tartrazine, sunset yellow FCF of curde propolis and commerical products, tartrazine was effect. The total phenolic average concentration of crude propolis was 1.64％, the total phenolic average concentration of commerical products was 1.64％. The lead content of curde propolis showed, especially high in the percentage of China (Hwa-pei area) was 163.33 ppm, the lead concent of 12 commerical products was 0 ppm, but CE-1 was 11.67 ppm. In the antimicrobial characteristics for Staphylococcus aureus showed, really not possess the similar effect of the sources of propolis and ethanolic extract products better than nonethanolic extract products. (Keywords：propolis, resin, flavonoids, phenolic, antimicrobial, analysis)

Keywords : propolis ; resin ; flavonoids ; phenolic ; antimicrobial ; analysis

Table of Contents

之類黃酮、黃酮醇與黃酮類含量 59 表十五、不同產地來源市售蜂膠產品萃取液（非乙醇類）、藥錠、膠囊產品之總酚含量 63 表十六、不同劑型市售蜂膠產品萃取液（非乙醇類）、藥錠、膠囊產品之總酚含量 64 表十七、市售蜂膠萃取液之鉛含量 65 表十八、不同產地來源蜂膠原膠對金黃色葡萄球菌之抑菌活性 69 表十九、不同產地來源市售蜂膠乙醇萃取液對金黃色葡萄球菌之 抑菌活性 70 表二十、不同產地來源市售蜂膠產品萃取液（非乙醇類）、藥錠、 膠囊產品對金黃色葡萄球菌之抑菌活性 71

REFERENCES

參考文獻 1. 于先覺。1997。蜂針配合蜂膠、中藥治療肺氣腫臨床觀察。中國養蜂 138（1）:19。 2. 川村賢司。1996。蜂膠之有效成分探討。蜂膠健康讀本1（3）:22-24。 3. 中國國家標準。1986。飼料檢驗法（砷之測定）。總號2770-17，類號N4024-17。經濟部中央標準局。 4. 王南舟。1991。蜂膠丙二醇提取液局部麻醉作用的實驗研究。蜜蜂雜誌 5:5-6。 5. 王進崑和孫璐西。1993。檳榔嚼塊中酚類化合物之分析。中國農業化學會誌 31（5）:623-632。 6. 日本健康、營養食品協會。1995。蜂膠食品規格基準公告－蜂膠食品及加工食品規格基準。pp1-10 7. 史滿田和何妤。1998。蜂膠降血黏度濃縮液對自由基過氧化脂質及血液黏度的影響。中國養蜂 49（5）:33-34。 8. 白?龍井及石塚忠生編著。1999。蜂膠療效小百科。林瑞玉譯。世貿出版社。 9. 朱燕華。1998。類黃酮之介紹。食品工業月刊 9（30）:1-5。 10. 江文德。1998。簡介大豆中的異黃酮素。食品工業月刊 30（9）:6-12。 11. 何鎧光和陳裕文。1999。蜂產品的研究及食療。中華傳統獸醫學會會刊 3（1）:19-34。 12. 吳萍。1995。蜂膠對微生物抑制作用的試驗報告。蜜蜂雜誌 12:3-4。 13. 吳萍和李正鵬。1996。蜂膠加多菌靈作為防污劑在平菇、金針菇、香菇中的應用。生物學雜誌 4:34-35。 14. 吳粹文、張復興、方兵兵、李熠、丁烈佛和張國南。1997。真理蜂膠營養液抗疲勞作用實驗研究。中國養蜂 6:9-10。 15. 呂澤田、姜得勇及田惠爭。1999。蜂膠中黃酮類化合物抑制腫瘤作用的試驗與應用。蜜蜂雜誌 3:8-10。 16. 宋心仿和邵有全。1999。蜂膠製劑保鮮效果對比試驗報告。中國養蜂 50（2）:6。 17. 李文源和陳莉萍。1992。蜂膠酊治療外耳道炎的臨床和實驗觀察。蜜蜂雜誌 2:3-4。 18. 李素菁。1999。蜂膠之抗氧化性及其類黃酮物質定量方法之探討。屏東科技大學食科技研究所 碩士論文。 19. 李榮明、陳宗欣、李秉鈺。1995。蜂膠臨床應用之展望。藥學雜誌 11（3）:103-108。 20. 李樹榮、尹培輝和王豔芬。1997。蜂膠制劑對羊鼻繩幼蟲禽體殺滅試驗。養蜂科技 6:5-6。 21. 李錦楓。1990。蜂產品之加工、檢驗與利用。中華昆蟲特刊 5:131-133。 22. 杜更堯。1990。蜂膠在禽蛋保鮮儲存中的應用初探。中國養蜂 3:28-29。 23. 房柱。1998a。蜂膠－黃酮化合物的寶庫。蜜蜂雜誌 9:11-12。 24. 林瑩禎。1996。保健食品市場近況。食品工業月刊 11:31-36。 25. 花美君、王金庸、張艷儒和李月賓。1991。蜂膠對高黏滯血症患者血液流變學的影響。蜜蜂雜誌 11-12:4-5。 26. 侯加強。1993。如何提高蜂膠酊的質量。養蜂科技 6:21。 27. 俞俊鰲。1996。蜂膠的研究。蜜蜂 3:23-25。 28. 施介人。1998。簡介槲皮酮。食品工業月刊 30（4）:53-61。 29. 茅力、楊森、陳景衡、金念組和魏國勤。1998。蜂膠醇溶液營養成分分析。南京醫科大學學報 18（6）:543-544。 30. 唐金賢、李素英、丁蓉民、史俊南和宋素萍。1995。用蜂膠制劑失活牙髓300例效果觀察。中國養蜂 4:3-4。 31. 唐傳和和孟岳成。1999。蜂膠的生理功能以及開發研究。食品工業科技 20（2）:30-32。 32. 徐明。1999。快速檢測蜂膠中重金屬鉛的方法。中國養蜂 50（1）:20-21。 33. 徐明和陳黎紅。1999。蜂膠中鉛的存在型式。蜜蜂雜誌 3:6-7。 34. 徐玲云和袁譯良。1989。蜂膠質量標準的探討。中國養蜂 2:6-8。 35. 徐誠、王國斌、陳恕仁、張恕峰和陳欣。1995。蜂膠酊抑制流感病實驗研究。蜜蜂雜誌 3:5-6。 36. 張嘉琪。2000。以HPLC法及光譜法測定蜂膠中之類黃銅物質。屏東科技大學食品科學研究所 碩士論文。 37. 曹治權主編。1992。微量元素與中醫藥。中國中醫藥出版社。北京。大陸。pp4。 38. 曹瑤和凌姬。1997。蜂膠中黃酮類化合物含量測定方法的改進。蜜蜂雜誌月刊 12:3。 39. 梁郁強、王俊、劉紅、韓云青及吳伯良。1999。中國養蜂 50（4）:5-6。 40. 許夏芬、張肇麟及朱燕華。2000。數種蔬菜中類黃酮含量及抗氧化分析。台灣農業化學與食品科學 38（5）:377-387。 41. 郭芳彬。1994。蜂膠在畜牧獸醫上的應用。養蜂科技 5:18-19。 42. 陳星山、劉安定、宮錫坤。1998。中藥抗菌作用研究。中醫藥學報 1:36-37。 43. 陳裕文和何鎧光。1999。蜂膠的生物效用。台灣養蜂業展望研討會專刊 70-82。 44. 陳福生。1991。蜂膠的抗菌性。中國養蜂 105（4）:41。 45. 陳澤隸。1992。蜂膠在鴨蛋保鮮儲存中的應用試驗。蜜蜂雜誌 8:5-6。 46. 彭和祿、李樹榮、易嘉賓和葛長榮。1993a。蜂膠、蜂花粉對豬肉質影響的研究。養蜂科技 4:9-10。 47. 彭和祿、李樹榮和李雲川。1992。蜂膠混懸劑皮下注射對小白鼠增重的試驗。養蜂科技 2:6-7。 48. 彭和祿、李樹榮和易嘉賓。1993b。蜂膠殘渣制劑對育肥豬增重效果的觀察。養蜂科技 2:6-7。 49. 彭增起、牛文娟和常彥紅。1996。蜂膠中的類黃酮及類黃酮的保健作用。中國養蜂 135（4）:14-15。 50. 彭增起、崔同和霍君生。1995。蜂膠精加工工藝。中國養蜂 127（2）:16。 51. 童麗霞。2000。蜂膠抗氧化性之研究。中山醫學院營養科學研究所 碩士論文。 52. 費經歐。1998。複方蜂膠酊治療68例肛門濕疹臨床體會。蜜蜂雜誌月刊 1:9。 53. 黃文誠。1997。蜂膠對小白鼠腹水癌的抗腫瘤作用。蜜蜂雜誌 1:7。 54. 黃堅。1998。開發蜂膠產品前景十分廣闊。養蜂科技 3:30-32。 55. 黃榮茂、王禹文、林聖富和楊得仁。1992。化學化工百科辭典。曉園。台北。台灣。pp433，729-730。 56. 黃衛平和劉放。1998。蜂膠的研究進展。養蜂科技5:6-11。 57. 楊朝環。1999。蜂產品民間應用法。蜜蜂雜誌月刊 6:14。 58. 蜂膠乙醇萃取液與加工食品的自由基準。1996。蜂膠規格成分含有量試驗方法。認證編號AA016623 日本蜂膠協議會。 59. 解玉軫。1999。不同種類蜂膠組成分分析及抗生特性研究。大葉大學食品工程研究所 碩士論文。 60. 廖大昆。1999。用蜂膠治療高脂血症。蜜蜂雜誌月刊 3:7。 61. 劉淑珍。1991。蜂膠是萬金油還是一種法定藥品。蜜蜂雜誌月刊3:11。 62. 劉富海。1999。中國農科院蜜蜂研究所。個人通訊。 63. 滕冰及吳宗璞。1999。大豆種粒中總多酚含量的分析方法。大豆科學 18（3）:265-268。 64. 蕭鳳岐。1996。蜂膠生物效用的利用。食品資訊 122:41-45。 65. 賴滋漢和賴業超。1994。食品科技辭典。富林。台中。台灣。pp1090，1091。 66. 遲家平、薛秉文和韓守智。1994。遼西蜂膠揮發物成分的研究。蜜蜂雜誌月刊 10:5-7。 67. 遲錫增主編。1997。微量元素與人體健康。化學工業出版社。北京。大陸。pp2。 68. 駱尚驊。1999。蜂膠質量控制的幾個指標和檢測方法。中國養蜂 50（3）:31。 69. 顏貽本。1985。蜂膠對血小板凝聚的體外試驗。蜜蜂 85（3）:19。 70. 顏貽本。1996。蜂膠對血小板凝聚的體外效應。蜜蜂 3:19。 71. 羅子程。2000。蜂膠原膠及市售產品之品質分析與抑菌活性。大葉大學食品工程研究所 碩

士論文。 72. 蘇華。1996。蜂膠在農產品防腐保鮮中的應用概況。蜜蜂雜誌 12:5-6。 73. 饒如龍。1988。蜂膠與其對糖尿病和癌症的醫療作用。中國養蜂 6:42-43。 74. 龔薇。1994。蜂膠治療昆明犬腸炎有奇效。養蜂科技 3:23-25。 75. 龔薇。1995。蜂膠在農牧業中的利用。養蜂科技 1:25-26。 76. Akimoto, K., Asami, S., Tanaka, T., Shimizu, S., Sugano, M. and Yamada, H. 1996. Antioxidant activity of sesamin on NADPH-dependent lipid peroxidation in liver microsomes. In: Natural Antioxidants and Food Quality in Atherosclerosis and Cancer Prevention. (Kumpulainen, J. T. and Salonen, J. T., eds). pp241-255. UK. 77. Albrecht, M., Frerick, H., Kuhn, U. and Strenge-Hesse, A. 1992. Therapy of toxic liver disease with Legalon. Z. Klin.Med. (Berlin). 47(2): 87-92. 78. Amoros, M., Lurton, E., Boustie, J., Girre, L., Sauvager, F. and Cormier, M. 1994. Comparison of the anti-herpes simplex virus activities of propolis and 3-methyl-2-enyl caffeate. J. Nat. Pro. 57(5): 644-647. 79. Amoros, M., Simoes, C. M., Girre, L., Sauvager, F. and Cormier, M. 1992. Synergistic effect of flavones and flavonols aginst herpes simplex virus type 1 in cell culture. Comparison with the antiviral activity of propolis. J. NAT. Pro. 55(12): 1732-1740. 80. Asaka, Y., Ohsaki, A., Kubota, T., Matsukawa, Y., Satomi, Y. and Nishino, H. 1992. 5,7,3',4'-Tetrahydroxy-3-methoxyflavone, a potent anti-tumor promoter isolated from Gnaphalium indicum. J. Kyo Preectural Univ. of Med. 101(4): 353-359. 81. Bankova, R., Christov, R., Stoev, G. and Popov, S. 1992a. Determination of phenolics from propolis by capillary gas chromatography. J. Chromatogr. 607: 150-153. 82. Bankova, V. G., Dyulgerov, A., Popov, S., Evstatieva, L., Kuleva, L., Pureb, O. and Zamjansan, Z. 1992b. Propolis produced in Bulgaria and Mongolia: phenolic compounds and plant origin. Apidologie. 23: 79-85. 83. Bankova, V. S., Christov, R. S. and Tejera, A. D. 1998. Lingans and other constituents of propolis from Canary Islands. Phytochem. 49(5): 411-415. 84. Bankova, V. S., Popov, S. S. and Marekov, N. L. 1983. A study on flavonoids of propolis. J. Nat. Pro. 46: 471-474. 85. Bankova, V. S., Popov, S. S. and Marekov, N. L. 1989. Isopentenyl cinnamates from poplar buds and propolis. Phytochem. 28(3): 871-873. 86. Bankova, V., Boudourova-Krasteva, G., Popov, S.,. Sforcin, J. M. and Cunlia Funari, S. R. 1998. Seasonal variations of the chemical composition of Brazilian propolis. Apidologie. 29: 361 -367. 87. Bankova, V., Christov, R., Popov, S., Marcucci, M. C., Tsvetkova, I. and Kujumgiev, A. 1999. Antibacterial activity of essential oils from Brazilian propolis. Fioterapia. 70: 190-193. 88. Bankova, V., Christov, R., Stoev, G. and Popov. S. 1992. Determination of phenolics from propolis by capillary gas Chromatography. J. Chromatogr. 607: 150-153. 89. Bankova, V., Marcucci, M. C., Simova, S., Nikolova, N., Kujumgiev, A. and Popov, S. 1996. Antibacterial diterpenic acids from Brazilian propolis. Z. Naturforsch. 51(5-6): 277-280. 90. Banskota, A. H., Tezuka, Y., Midorikawa, K. and Kadota, S. 2000. Two novel cytotoxic benzofuran dervatives from brazilian propolis. J. Nat. Pro. 63(9): 1277-1279. 91. Barth, O. M. 1998. Pollen analysis of Brazilian propolis. Grana 37(2): 97-101. 92. Basnet, P., Matsuno, T. and Neidlein, R. 1997. Potent free radical scavenging activity of propol isolated Brazilian propolis. Z. Naturforsch. 52(11-12): 828-838. 93. Basnet, P., Matsushige, K., Hase, K., Kadota, S. and Namba, T. 1996. Potent antihepatotoxic activity of dicaffeoyl quinic acids from propolis. Biol. Pharm. Bull. 19(4): 655-657. 94. Basnet, P., Matsushing, K., Kadota, S. and Namba, T. 1996. Four-di-O-caffeoyl quinic acid derivatives from propolis. Poten hepatoprotective activity in experimental liver injury models. Biol. Pharm. Bull. 19(11): 1479-1484. 95. Bernd, K. 1985. Plant sources of propolis. Bee World. 66: 136-139. 96. Bonvehi, J. S. and Coll, F. V. V. 1994. Phenolic composition of propolis from China and South America. Z. fur Naturforsch, section C. 49(11/12):712-718. 97. Brasseur, T. 1989. Anti-inflammatory properties of flavonoids. J. Phare. Belg. 44: 235-241. 98. Brinkworth, R. I., Stoermer, M. J. and Fairlie, D. P. 1992. Flavones are inhibitors of HIV-1 proteinase. Biochem. Biophys. Res. Com. 188(2): 631-637. 99. Brown, R. 1989. Hive products: pollen, propolis and royal jelly. Bee World. 70: 109-117. 100.Buckshee, K., Takkar, D. and Aggarwal, N. 1997. Micronized flavonoid therapy in internal hemorrhoids of pregnancy. J. Gynecolo and Obstetrics. 57(2): 145- 151 . 101.Burdock, G. A. 1998. Review of the biological properties and toxicity of bee propolis. Food Chem. Toxicol. 36(4): 347-363. 102.Chiao, C., Carothers, A. M., Gunberger, D., Solomon, G., Preston, G. A. and Barrett, J. C. 1995. Apoptosis and altered redox state induced by caffeic acid phenethyl ester (CAPE) in transformed rat fibroblast cells. Cancer Res. 55(16): 3576-3583. 103.Cholbi, M. R., Paya, M. and Alcaraz, M. J. 1991. Inhibitory effects of phenolic compounds on carbon tetrachloride induced microsomal lipid peroxidation. Exp. 47(2): 195-199. 104.Chopra, S., Pillai, K. K., Husain, Z. and Giri, DK. 1995. Propolis protects against doxorubicin-induced myocardiopathy in rats. Exp. and Mol. Path. 62: 190-198. 105.Christov, R., Bankova, V., Tsvetkova, I., Kujumgiev, A. and Tejera, A. D. 1999. Antibacterial furofuran lignans from Canary Islands propolis. Fitoterapia. 70: 82-92. 106.Christov, R., Bankova, V., Tsvetkova, I., Kujumgiev, A. and Tejera, A. D. 1999. Antibacterial furofuran lignans from Canary Islands propolis. Fitoterapia. 70: 82-92. 107.Crane, E. 1996. The past and present importance of bee products to men. Bee Products.1: 1-14. 108.Crozier, A., Lean, M. E. J., McDonald, M. S. and Black, C. 1997. Quantitative analysis of the flavonoid content of commercial tomatoes, onions, lettuce and celery. J. Agric. Food Chem. 45: 590-595. 109.Debiaggi, M., Tateo, F., Pagani, L., Luini, M. and Romero, E. 1990. Effects of proolis flavonoids on virus infectivity and replication. Microbiol. 13(3): 207-213. 110.Dobrowolski Jan. W., Vohora, S. B., Sharma Kalpana., Shaukat Shan. A., Naqvi, S. A. H. and Dandiya, P. C. 1991. Antibacterial, antifungal, antiamoebic, antiinflammatory and antipyretic studies on propolis bee products. J. Ethnopharmacology. 35: 77-82. 111.Dumitrescu, M., Crisan, I. and Esanu, V. 1993. The mechanism of the antiherpetic action of an aqueous propolis extract. II. The action of the lectins of an aqueous propolis extract. Rev. Roum. Virol. 44(1-2): 49-54. (Freench) 112.Esanu, V., Prahoveanu, E., Crisan, I. and Cioca, A. 1981. The effect of an aqueous propolis extract, of rutin and of a rutin-quercetin mixture on experimental influenzal virus infection in mice. Virplpgie. 32(3): 213-215. 113.Ewald, C., Fjelkner-Modig, S., Johansson, K. and Sjoholm, I. 1999. Effect of processing on major flavonoids in processed onions, green beans and peas. Food Chem. 64: 231-235. 114.Frenkel, K., Wei, H., Bhimani, R., Ye, J., Zadunaisky, J., Huang, M. T., Ferraro, T., Conney, A. H. and Grunberger, D. 1993. Inhibition of tumor promoter-mediated processes in mouse skin and bovine lens by caffeic acid phenethyl ester. Cancer Res. 53(6): 1255- 1261. 115.Fujimoto, T. 1992. Qualitative and quantitative characteristics of propolis and its products. Honeybee Sci. 13(4): 145-150. 116.Ghisalberti, E. L. 1979. Propolis: a review. Bee World. 60: 59-84. 117.Ghisalberti, E. L., Jefferies, P. R., Lanteri, R. and Matisons, L. 1978. Constituents of propolis, Experientia.

34(2): 157-158. 118.Grange, J. M. and Davey, R. W. 1990. Antibacterial propoerties of propolis (bee glue). J. R. Soc. Med. 83(3): 159-160. 119.Greenaway, W., Scaysbrook, T. and Whatley, F. R. 1990. The composition and plant origins of propolis: a report of work at Oxford. Bee World 71: 107-118. 120.Greenway, W., May, J., Scaysbrook, T. and Whatley, F. R. 1991. Identification by gas chromatography-mass spectrometry of 150 compounds in propolis. Z. fur Naturforschung. 46c: 111-121. 121.Greenway, W., Scaysbrook, T. and Whatley, F. R. 1987. The analysis of bud exudate of Polulus X euramericana, and of propolis, by gas chromatography-mass spectrometry. Proc. R. Soc. Lond.B. 232, 249-272. 122.Grunberger, D., Banerjee, R., Eisinger, K., Oltz, E. M., Efros, L., Caldwell, M., Esteve, V. and Nakanishi, K. 1988. Preferential cytoxicity on tumor cells by caffeic acid phenethyl ester isolated from propolis. Experientia 4(3): 230-232. 123.Guarini, L., Su, Z. Z., Zucker, S., Lin, J., Grunberger, D. and Fisher, P. B. 1992. Growth inhibition and modulation of antigenic phenotype in human melanoma and glioblastoma multiforme cells by caffeic acid phenethyl ester. Cell Mol. Biol. 38(5): 513-527. 124.Gyorgy, I., Antus, S., Blazovics, A. and Foldiak, G. 1992. Substituent effects in the free radical reactions of silybin:Radiation-induced oxidation of the flavonoid at neutral pH. Intern. J. Radiation Bio. 61(5): 603-609. 125.Hertog, M. G. L., Hollman, P. C. H. and Katan, M. B. 1992. Content of Potentially anticarcinogenic flavonoids of 28 vegetables and 9 fruits commonly consumedin the Netherlands. J. Agric. Food Chem. 40: 2379-2383. 126.Hirota, M., Matsuno, T., Fujiwara, T., Sugiyama, H. and Mineshita, S. 2000. Enhanced cytotoxicity in a Z-photoisomr of a benzopyran derivative of propolis. J. Nat. Prod. 63(3): 366-370. 127.Hollman, P. C. H., Hertog. M. G. L. and Katn, M. B. 1996. Analysis and health effects of flavonoids. Food Chem. 57: 43-46. 128.Huang, M. T., Ma, W., Yen, P., Xie, J. G., Han, J., Frenkel, K., Grunberger, D. and Conney, A. H. 1996. Inhibitory effcts of caffeic acid phenethyl ester (CAPE) on 12-O-tetradecanoylphorbol-13-acetate-induced tumor promotion in mouse skin and the synyhesis of DNA, RNA and protein in HeLa cells. Carcinogenesis. 17(4): 761-765. 129.Iannuzzi, J. 1983. Propolis : the most mysterious hive element. Amer. Bee. J. 123: 631-633. 130.Iannuzzi, J. 1990 . America`s propolis king. Gleaning Bee Cult. 188: 480-481. 131.Ivanovsaka, N. D., Dimov, V. B., Bankova, V. S. and Popov, S. S. 1995b. Immunomodulatory action of propolis. VI. Influence of a water soluble derivative on complement activity in vivo. J. Ethnopharmacol. 47:145-147. 132.Ivanovsaka, N. D., Dimov, V. B.,Pavlova, S., Bankova, V. S. and Popov, S .S. 1995a. Immunomodulatory action of propolis. V. Anticomplementary activity of a water-soluble derivative. J. Ethnopharmacol. 47: 135-143. 133.Jaiswal, A. K., Venugopal, R., Mucha, J., Carothers, A. M. and Grunberger, D. 1997. Caffeic acid phenethyl ester stimulates human antioxidant response element-mediated expression of the NAD(P)H: quinone oxidoreductase (NQO1) gebe. Cancer Res. 57(3): 440-446. 134.Jolly, B.G. 1978. Propolis varnish for violins. Bee World. 59:157-161. 135.Kamei, H., Koide, T., Kojimam, T., Hasegawa, M., Terabe, K., Umeda T. and Hashimoto Y. 1996. Flavonoid mediated tumor growth suppression demonstrated by in vivo study. Cancer Biotherapy and Radiopharmaceuticals. 11(3): 193-196. 136.Khayyal, M. T., et-ghazaly, M. A., el-khatib, A. S. 1993. Mechanisms involved in the anti-inflammatory effect of propolis extract. Drug Exp Clin Res. 19(5): 197-203. 137.Kimoto, T., Arai, S., Aga, M., Hanaya, T., Kohguchi, M., Nomura, Y. and Kurimoto, M. 1996. Cell cycle and apoptosis in cancer induced by the artepillin C extracted from Brazilian propolis. Gan To Kagaku Ryoho. 23(13): 1855-1859. (Japanese). 138.Kimoto, T., Arai, S., Kohguchi, M., Aga, M., Nomura, Y., Micallef, MJ., Kurimoto, M. and Mito, K. 1998. Apoptosis and suppression of tumor growth by artepillin C extracted from Brazilian propolis. Cancer Detect Prev. 22(6): 506-515. 139.Koltuksuz, U., Ozen, S., Uz., E., Aydinc, M., Karaman, A., Gultek, A., Akyol, O., Gursoy, M. H. and Aydin, E. 1999. Caffeic acid phenethyl ester prevents intestinal reperfusion injury in rats. J. Pediatr. Surg. 34(10): 1458-1462. 140.Koo, H., Gomes, B. P. F. A., Rosalen, P. L., Ambrosano, G. M. B., Park, Y. K. and Cury, J. A. 2000. In vutro antimicrobial activity of propolis and Arnica montana againet oral pathogens. Arch. Oral Biol. 45: 141-148. 141.Kosonocka, L. 1990. Propolis-snake oil or legitimate medicine. Amer. Bee. J. 130: 451-452. 142.Krol, W., Czuba, Z., Scheller, S., Gabrys, J., Grabiec, S. and Shani, J. 1990. Anti-oxidant property of ethanolic extract of propolis (EEP) as evaluated by inhibition the chemiluminescence oxidation of luminol. Biochem. Int. 21(4)593-597. 143.Krol, W., Scheller, S., Czuba, Z., Matsuno, T., Zydowicz, G., Shani, J. and Mos, M. 1996. Inhibition of neutrophils' chemiluminescence by ethanol extract of propolis (EEP) and its phenolic components. J. Ethanopharmacol. 55(1): 19-25. 144.Krol, W., Scheller, S., Shani, J., Pietsz, G. and Czuba, Z. 1993. Synergistic effect of ethanolic extract of propolis and antibiotics on the growth of Staphylococcus aureus. Arzneimittelforschung. 43(5): 607-609. 146.Kujumgiev, A., Tsvetkova, I., Serkedjieva, Yu., Bankova, V., Christov, R. and Popov, S. 1999. Antibacterial, antifungal and antiviral activity of propolisof different geographic origin. J. of Ethnopharmacology. 64: 235-240. 147.Kujumgiev, A., Tsvetkova, I., Serkedjieva, Yu., Bankova, V., Christov, R. and Popov, S. 1999. Antibacterial, antifungal and antiviral activity of propolisof different geographic origin. J. of Ethnopharmacology. 64: 235-240. 148.Lindenfelser, L. A. 1968. Lnvivo activity of propolis against Bacillus larvae. J. Invert. Path. 12: 129-131. 149.Lowe, D. G. 1980. Propolis substitutes.Bee World.61: 120-121. 150.Lu, H. Q., Niggemann, B. and Zanker, K. S. 1996. Suppression of the proliferation and migration of oncogenic ras-dependent cell lines, cultured in a three-dimensional collagen matrix, by flavonoid-structured molecules. J. of Cancer Res and Clin Oncol. 122(6): 335-342. 151.Macedo, F. A. N. 1996. Heavy metals in propolis:Practical and sample proceduce the level in the Brazilian propolis. Bee products.28: 231-238. 152.Maciejewice, W., Daniewski, M., Mielniczuk, Z., Suprynoyowica, Z. 1982. Gas Chromatography-Mass Spactrometry Investigation of Propolis. Analysis of ?-Steroids. Acta. Polon. Pharm. XXXIX Nr. 4: 277-279. 153.Mahram, L. G., el-kliatib, A. S., Agha, A. M., and Khayyal, M. T.1996. The protective effect of aqueous propolis extract on isolated rat hepatocytes against carbon tetrachloride toxicity. Drug Exp. Clin. Res. 22(6): 309-316. 154.Marcucci, M. C. 1995. Propolis: chemical composition, biological properties and therapeutic activity. Apidologie. 26: 83-99. 155.Marinescu, I. and Tamas, M. 1980. Poplar buds-a source of propolis. Apiacta 15: 121-26. 156.Matsuno, T., Jung, S. K., Matsumoto, Y., Saito, M. and Morikawa, J. 1997a. Preferential cytotoxicity to tumor cells of 3,5-diprenyl-4-hydroxycinnamic acid (artepillin C) isolated from proolis. Anticancer Res. 17(15A): 3565-3568. 157.Matsuno, T., Matsumoto, Y., Saito, M. and Morikawa, J. 1997b. Isolation and characterization of cytotoxic diterpenoid isomers from propolis. Z. Naturforsch. 52(9-10):

702-704. 158.McGregor, S. E. 1952. Collection and utilization of propolis and pollen by caged honey bee colonies. Amer.Bee. J. 29: 20-21. 159.Menezes, H., Bacci Jr, M., Oliverira, S. D. and Pagnocca, F. C. 1997. Antibacterial properties of propolis and products containing propolis from Brazil. Apidologie. 28: 71-76. 160.Meresta, L. and Meresta, T. 1985. Antibacterial activity of flavonoid compounds of propolis, occurring in flora in Poland. Med. Weter 41:489-492. 161.Metzner, J., Bekemeier, H. and Weber, F. G. 1982. Influence of flavonoids on capillary permeability, carrageenin edema and histamine and PGE2 spasms. Agents Actions Suppl. 10: 129-134. 162.Mirzoeva, O. K. and Calder, P. C. 1996. The effect of propolis and its components on eicosanoid production during the inflammatory response. Prostaglandins Leukot Essent Fatty Acids. 55(6): 441 -449. 163.Mirzoeva, O. K., Sud ' ina, G. F., Pushkareva, M. A., Korshunova, G. A., Sumbatian, N. V. and Varfolomeev, S. D. 1995. Lipophilic derivatives of caffeic acid as lipoxygenase inhibitors with antioxidant properties. Bioorg. Khim. 21(2): 143-151.(Russian). 164.Mirzoeva, O. K., Yaqoob, P., Knox, K. A. and Calder, P. C. 1996. Inhibition of ICE-family cysteine proteases rescues murine lymphocytes from lipoxygenase inhibitor-induced apoptosis. FEBS Lett. 396(2-3): 266-270. 165.Misic, V., Ondrias, K., Gergel, D., Bullova, D., Suchy, V. and Nagy, M. 1991. Lipid peroxidation of lecithin liposomes depressed by some constituents of propolis. Fttoterapia. 7(3): 215-220. 166.Mitamura, T., Matsuno, T., Sakamoto, S., Maemura, M., Kudo, H., Suzuki, S., Kuwa, K., Yosh.imura, S., Sassa, S., Nakavama, T. and Nagasawa, H. 1996. Effects of a new clerodane diterpenoid isolated from propolis on chemically induced skin tumors in mmice. Anticancer Res. 16(5A): 2669-2672. 167.Miyakado, V. and Sulimanovic, D. 1982. Action of propolis solutions on Bacillus larvae. Apiacta. 17: 16-20. 168.Morales, W. F. and Garbarino, J. L. 1996. Clinical evaluation of a new hypoallergic formula of propolis in dressings. Bee Products. 13: 101-106. 169.Moreno, M. I., Isla, M. I., Sampietro, A. R. and Vattuone, M. A. 2000. Comparison of the free redical-scavenging activity of propolis from several regions of Argentina. J. Ethnopharmacol. 71(1-2): 109-114. 170.Na, H. K., Wilson, M. R., Kang, K. S., Chang, C. C., Grunberger, D. and Trosko, J. E. 2000. Restoration of gap junctional intercellular communication by caffeic acid phenethyl ester (CAPE) in a ras-transformed rat liver epithelial cell line. Cancer Lett. 157(1): 31-38. 171.Natarajan, K., Singh, S., Burke, T. R. Jr., Grunberger, D. and Aggarwal, B. B. 1996. Caffeic acid phenethyl ester is a potent and specific inhibitor of activation of nuclear transcription factor NF-kappa B. Proc. Natl. Acad. Sci. U. S. A. 93(17): 9090-9095. 172.Nieva Moreno, M. I., Isla, M. I., Cudmani, N. G., Vattuone, M. A. and Sampietro, A. R. 1999. Screening of antibacterial activity of Amaicha del Valle (Tucuman, Argentina) propolis. J. of ethnopharmacology. 68: 97-102. 173.Nivia, M. F. A. 1996. Heavy metals in propolis : pricaical and simple procedures to reduce the lead leavel in the Brazilian propolis. Bee Products. 28: 231-238. 174.Omori, H., Nio, Y., Minari, Y., Takeda, H., Sato, Y., Song, M. M., Hirahara, N., Sumi, S. and Tamura, K. 1996. Anti-tumor effects of quinolinone derivatives, vesnarinone (OPC-8212) on human pancreas cancer cell lines. J. Jpn. Soc. Cancer Therapy. 31(7): 446-455. 175.Paintz, M. and Metzner, J. 1979. Zur lokalanasthetischen Wirkung von Propolis und einigen Inhaltsstoffen. Pharmazie. 34: 839-841. 176.Par, A. 1992. Pathogenesis and management of alcoholic liver injury. Acta Physiologica Hungarica. 80(1-4): 325 – 350. 177.Parrk, Y. K., Koo, M. H., Ikegaki, M. and Contado, J. L. 1997. Comparison of the flavonoid aglycone contents of Apis mellifera propolis from various regions of Brazil. Arq. Biol. Tecnol. 40(1): 97-106. 178.Pascual, C., Gonzalez, R., Torricella, R. G.. 1994. Scavenging action of propolis extract against oxygen radicals. J. Ethiiopharmacol. 41(1-2): 9-13. 179.Pecking, A. P. 1995. Evaluation of lymphoscintigraphy of the effect of a micronized flavonoid fraction (Daflon 500 mg) in the treatment of upper limb lymphedema. International Angiology 14: 39-43. 180.Pepeljnjak, S., Jalsenjak, I. and Maysinger, D. 1982. Growth inhibition of Bacillus subtilis and composition of various propolis extracts. Pharmazie. 37(12): 864-865. 181.Pepeljnjak, S., Jalsenjak, I. and Maysinger, D. 1985. Flavonoid content in propolis extracts and growth inhibition of Bacillus subtilis. Pharmazie 40(2): 122-123. 182.Qiao, Z. and Chen, R. 1991. Isolation and identification of antibiotic constituents of propolis from Henan. Chung Kuo Chung Yao Tsa Chih 16(8): 481-482, 512. (Chinese). 183.Rao, C. V., Desai, D., Kaul, B., Amin, S. and Reddy, B. S. 1992. Effect of cafffeic acid esters on carcinogen-induced mutagenicity and human colon adenocarcinoma cell growth. Chem. Biol. Interact. 84(3): 277-290. 184.Rao, C. V., Desai, D., Rivenson, A., Simi, B., Amin, S. and Reddy, B. S. 1995. Chemoprevention of colon carcinogenesis by phenylethyl-3-methylcaffeate. Cancer Res. 55(11): 2310-2315. 185.Rao, C. V., Desai, D., Simi, B., Kulkarni, N., Amin, S. and Reddy, B. S. 1993. Inhibitory effect of caffeic acid esters on azoxymethane-induced biochemical changes and aberrant crypt foci formation in rat colon. Cancer Res. 53(18): 4182-4188. 186.Rapta, P., Misik, V., Stasko, A. and Vrabel, I. 1995. Redox intermediates of flavonoids and caffeic acid esters from propolis: an EPR spectroscopy and cyclic voltammetry syudy. Free Radic. Biol. Med. 18(5): 901-908. 187.Rodriguez, E. G., Abellan, G. B. and Villanueva, M. T. O. 1999. Macroelements in dietetic products containing propolis. Food Chemistry. 66:15-19. 188.Roger, C. R. 1988. The nutritional incidence of flavonoids:some physiological and metabolic considerations. Experientia. 44: 725-733. 189.Rubio, O. C., Cuellar, A., Rojas, N., Castro, H. V., Rastrelli, L., and Aquino, R. 1999. A polyisoprenylated benzophenone from Cuban propolis. J. Nat. Prod. 62(7): 1013-1015. 190.Scheer, J. F. and Writer, C. 1998. Products of the hive-sticky, sweet and healthful. Better Nutrition. 60-64. 191.Scheller, S., Gazda, G. and Pietsz, G., Gabrys, J., Szumlas, J., Eckert, L. and Shani, J. 1988. The ability of ethanolic extract of propolis to stimulate plaque formation in immunized mouse spleen cells. Pharmacological Res. Communications. 20(4): 323-328. 192.Schmidt, J. O. 1996. Bee products:chemical composition and application. Bee products. 2: 15-26. 193.Serkedjieva, J., Manolova, N., Bankova, V. 1992. Anti-influenza virus effect of some propolis constituents and their analogues(esters of substituted cinnamic acids). 194.Sforcin, J. M., Fernandes Jr. A., Lopes, C. A. M., Bankova, V. and Funari, S. R. C. 2000. Seasonal effects on Brazilian propolis antibacterial activity. J. Ethnopharmacology. 73: 243-249. 195.Shieh, Y. H., Lin, Y. C., Lin, S. C. and Lin, Y. H. 1997. Protective and therapeutic effects of propolis on carbon tetrachloride induced liver injuries.內科學誌 8: 137-142. 196.Sidwell, R. W., Huffman, J. H., Moscon, B. J. and Warren, R. P. 1994. Influenza virus-inhibitory effects of intraperitoneally and aerosol-administered SP-303, a plant flavonoid. Chemother. 40(1): 42-50. 197.Siess, M. H., Bon, A. M. L., Canivenc-Lavier, M. C., Amiot, M. J., Sabatier, S., Aubert, S. Y. and Suschetet, M. 1996. Flavonoids of honey and propolis: Characterization and effects on hepatic drug-metabolizing

enzymes and benzo[α]pyrene-DNA binding in rats. J. Agric. Food Chem. 44: 2297-2301. 198.Singh, Z. 1972. Propolis collection and its use.Indian Bee J. 34: 11-19. 199.Sorkun, K. Bozcuk, S., Gomurgen, A. N. and Tekin, F. 1996. An inhibitory effect of propolis on germination and cell division in the root tips of wheat seedings. Bee Products. 17: 129-136. 200.Strehl, E., Volpert, R. and Elstner, E. F. 1994. Biochemical activities of propolis extracts. III. Inhibition of dihydrofolate reductase. Z. Naturforsch. 49(l-2):3 9-43. 201.Su, Z. Z., Grunberger, D. and Fisher, P. B. 1991. Suppression of adenovirus type 5 E1A-mediated transformation and expression of the transformed phenotype by caffeic acid phenethyl ester (CAPE). Mol. Carcinog. 4(3): 231-242. 202.Su, Z. Z., Lin, J., Grunberger, D. and Fisher, P. B. 1994. Growth suppression and toxicity induced by caffeic acid phenethyl ester (CAPE) in type 5 adenovirus-transformed rat embryo cells correlate directly with transformation progression. Cancer Res. 54(7): 1865-1870. 203.Su, Z. Z., Lin, J., Prewett, M., Goldstein, N. I. and Fisher, P. B. 1995. Apoptosis mediates the selective toxicity of caffeic acid phenethyl ester (CAPE) toward oncogene-transformed rat embryo fibroblast cells. Anticancer Res. 15(5B): 1841-1848. 204.Sud'ina, G. F., Mirzoeva, O. K., Pushkareva, M. A., Korshunova, G. A., Sumbatyan, N. V. and Varfolomeev, S. D. 1993. Caffeic acid phenethyl ester as a lipoxygenase inhibitor with antioxidant properties. FEBS Lett. 329(1-2): 21-24. 205.Tatefuji, T., Izumi, N., Ohta, T., Arai, S., Ikeda, M. and Kurimoto, M. 1996. Isolation and identification of compounds from Brazilian propolis which enhance macrophage spreading and mobility. Biol. Pharm. Bull. 19(7): 966-970. 206.Tatefuji, T., Yamauchi, H., Ikeda, M. Ando, S. and Kurimoto, M. 1993. Effect of propolis obtained in Brazil on infectivity of viruses. Shoyakugaku Zasshi 47: 60-64. 207.Tomas-Barbran, F. A., Cristina-Viguera, C., Vit-Olivier, P., Ferreres, F., Tomas-Lorente, F. 1993. Phytochemical evidence for the botanical origin of tropical propolis from venezuela. hytochemistry.34(1): 191-196. 208.Tosi, B., Donini, A., Romagnoli, C. and Bruni, A. 1996. Antimicrobial Activity of Some Commerical Extracts of Propolis Prepared with Different Solvents. Phytotherapy Res. 10:335-336. 209.Valcic, S., Montenegro, G., Mujica, A. M., Avila, G., Franzblau, S., Singh, M. P., Maiese, W. M. and Timmermann, B. N. 1999. Phytochemical, morphological and biological investigations of propolis from Central Chile. Z. Naturforsch. 54(5-6): 406-416. 210.Velikova, M., Bankova, V., Marcucci, M. C., Tsvetkova, I. and Kujumgiev, A. 2000a. Chemical composition and biological activity of propolis from Brazilian meliponinae. Z. Naturforsch. 55(9-10): 785-789. 211.Velikova, M., Bankova, V., Sorkun, K., Houcine, S., Tsvetkova, I. and Kujumgiev, A. 2000b. Propolis from the Mediterranean region: chemical composition and antimicrobial activity. Z. Naturforsch. 55(9-10): 790-793. 212.Vereckei, A., Feher, E., Gyorgy, I., Foldiak, G., Toth, M., ToncSey, H. and Feher, J. 1991. The possibilities of antioxidant therapy in the prevention of amiodarone side effects. Orvosi Hetilap 132(23):1265- 1268. 213.Walker, P. and Crane, E. 1987. Constituents of propolis. Apidologie 18(4): 327-334. 214.Wang, C. K. and Lee, W. H. 1996. Separation, characteristics and biological activities of phenolics in area fruit. J. of