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药用植物真菌毒素污染现状与控制策略

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摘　要: 药用植物被广泛用作家庭医疗和制药工业的原材料。随着消费量和种植面积的逐年增加,药用植物真菌毒素污染有加重的趋势。在生长、收获、处理和储存的过程中,药用植物都可能会受到各种霉菌的污染,从而导致其发霉并产生真菌毒素。加之缺乏对这些天然药品质量和毒性的有效监测,其安全性已经成为一个严重的公共卫生安全问题。本文对药用植物中主要的真菌毒素污染种类及其天然存在情况进行了综述,提出了真菌毒素污染防控措施,并对保障药品质量安全和人体健康采取的方法进行了展望。

关键词: 药用植物;真菌毒素;污染;控制

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Present situation and control strategy of mycotoxin contamination in medicinal plants

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Abstract: Medicinal plants are widely used as raw materials in the family medicine and pharmaceutical industries. However, with increases in the consumption and planting area of medicinal plants each year, mycotoxin contamination has also increased. During their growth, harvest, treatment, and storage, medicinal plants may be contaminated by various molds that can produce mycotoxins. Effective methods for monitoring the quality and toxicity of these natural drugs are lacking, leading to serious public health risks. In this study, the types and natural existence of mycotoxins in medicinal plants are reviewed, prevention and control measures of mycotoxin contamination are proposed, and methods for ensuring drug safety and human health are described.

Keywords: medicinal plant; mycotoxin; contamination; control

世界卫生组织将药用植物定义为具有某些特殊特性的植物,这些特性使其有资格成为药物和治疗剂,并被用于药用目的[1]。一种植物被指定为“药用”时,是指该植物可以作为一种治疗剂或药物或一种药物配方的有效活性成分[2]。植物药也被称为中草药,自古以来就被用于预防、治疗疾病和促进健康,几千年来在包括我国在内的世界各国的卫生问题中发挥了重要作用。在欠发达国家,由于缺乏现代医疗保健,使用草药非常普遍。如今,药用植物的消费正在增加,在过去的几十年中,药用植物的使用已经发展到全球。1995年,全球药用植物市场仅为170亿美元,最近的一项市场分析表明,到2023年药用植物市场份额将增加至1 110亿美元(复合年增长率为7%~8%)[3]。目前在世界范围内,药用植物被广泛用作家庭治疗和制药工业的原料[4]。据估计,全球约80%的人口在初级卫生保健的某些方面使用过草药[5],发展中国家约89%的人口使用过传统草药[6]。美国食品和药物管理局估计,现在全球25%~50%的药物来自不同的植物材料[7],每年有超过2万种不同的植物被用于药用植物治疗,随着全球药用植物使用的日益广

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泛，药用植物的质量控制问题已成为卫生当局和公众关切的热点[8]。

但是，市场上出售的植物药并不总是符合质量和安全标准，由于药用植物消费的大量增加，加之缺乏对这些天然产品使用、功效、毒性和质量的有效监测，其使用已成为一个公共卫生安全问题。目前，大量综述报道了植物药和相关产品中真菌毒素污染的发生情况，这些研究表明植物药中的霉菌毒素污染是一个十分严重的全球性问题。在生长、收获、处理、储存和分销过程中，药用植物均可能受到各种真菌的污染，从而导致药用植物的腐败和真菌毒素的产生[9-10]，这种情况在经济不发达、生产不规范的国家或地区表现得尤为严重。中草药使用量的增加可能导致人体真菌毒素摄入量的增加，因此，要严格防范真菌毒素污染药用植物可能导致的人类健康问题。

真菌毒素是由某些丝状真菌（霉菌）产生的有毒次级代谢产物，定义为经摄入、吸入或通过皮肤吸收可导致疾病或人类和动物死亡的真菌代谢物[11]。真菌毒素可引起多种毒性作用，其具有致癌、致畸、神经毒性和免疫毒性[12-13]。目前已经发现了400种不同的真菌毒素[14]，从人类和家畜健康的角度来看，最主要的真菌毒素为黄曲霉毒素、赭曲霉毒素、伏马菌素、玉米赤霉烯酮和呕吐毒素[15-17]。这些毒素通常以污染物的形式存在于不同的商品中，包括粮油作物、药用植物、坚果和香料等。迄今为止，在药用植物中共检测到40多种真菌毒素，这些产毒真菌包括曲霉属、青霉属、镰刀菌属和链霉菌属，有关报道显示，黄曲霉毒素、赭曲霉毒素、伏马菌素、毛癣菌素和玉米赤霉烯酮是植物药中最常见的污染真菌毒素，且已多次报道其浓度超过欧盟规定的监管水平[18]。土壤或植物中的产毒真菌均可以导致药用植物污染，除了真菌毒素本身的毒性作用外，植物药中真菌毒素的存在还可能会降低药效，导致药物间的相互作用，并可能加剧影响中草药安全性的副作用。

1 药用植物中的黄曲霉毒素污染

研究人员对多种药用植物中的黄曲霉毒素进行了分析。YANG等[19]在19个中草药样品中发现3个样品被黄曲霉毒素污染（32 $\mu g \cdot kg^{-1}$），并观察到其中板栗受污染水平最高。有研究采用高效液相色谱-串联质谱法对来自国内和国际供应商的174份药用植物样品进行了黄曲霉毒素总污染分析，其中15.5%的样品黄曲霉毒素检测呈阳性，且浓度高达290.8 $\mu g \cdot kg^{-1}$[20]。建议应该严格监测富含油脂的药用植物的真菌毒素含量，因为这些药用植物可能更有利于霉菌的生长。然而在另一项研究中，只有5.2%的药用植物样品被证实含有黄曲霉毒素[21]。HAN等[22]建立了一种免疫亲和提取-超高效液相色谱-串联质谱方法检测药用植物的黄曲霉毒素B_1、B_2、G_1和G_2的含量，该方法准确、灵敏，还可用于检测其他类似药用植物的复杂基质。

其他国家也有关于药用植物的黄曲霉毒素污染情况的报道。在印度的药用植物中经常检测到较高水平的黄曲霉毒素，印度当地仓库的37个样本中，30个样本被发现受到黄曲霉毒素污染[23]。在另一项研究中，药用植物中黄曲霉毒素B_1和桔霉素的含量分别为0.02~1.18 $\mu g \cdot g^{-1}$和0.01~0.76 $\mu g \cdot g^{-1}$[24]。对用于治疗肝病的草药进行检测的结果显示，在50个样本中，有46%的草药含有黄曲霉毒素，其中天门冬中黄曲霉毒素B_1含量最高，达到2.23 $\mu g \cdot g^{-1}$[25]，该结果令人震惊，因为发现用于治疗肝病的药物本身就含有大量的肝毒素。在土耳其，有报道显示无花果干中含有大量的黄曲霉毒素，对从当地不同商店和出口工厂收集的无花果干样本使用高效液相色谱进行检测：本地商店样品中黄曲霉毒素污染的发生率较高，219个样品中有47.5%的样品被污染，而2 461个出口样品中有23.6%的样品被污染；本地市场样品中黄曲霉毒素浓度高达267.48 $ng \cdot g^{-1}$，出口样品中黄曲霉毒素浓度高达278.04 $ng \cdot g^{-1}$[26]。土耳其的另一项研究也报道了无花果干中的黄曲霉毒素污染情况，黄曲霉毒素B_1的水平为112.3 $ng \cdot g^{-1}$，黄曲霉毒素B_2的水平为50.6 $ng \cdot g^{-1}$，黄曲霉毒素G_1的水平为61.4 $ng \cdot g^{-1}$[27]。HEPERKAN等[28]的研究显示，23%的无花果干样品中同时存在黄曲霉毒素和环吡嗪酸。IAMANAKA等[29]在58%的无花果干样本中检测到了0.3~2.0 $\mu g \cdot kg^{-1}$水平的黄曲霉毒素，其中一个样本中黄曲霉毒素B_1的浓度极高，达到1 500 $\mu g \cdot kg^{-1}$。一项对埃及31份药用植物样品进行检测的研究发现，29%的样品被确定含有黄曲霉毒素B_1，其平均浓度为49 $\mu g \cdot kg^{-1}$[30]。

尼日利亚、马来西亚、阿曼、印度尼西亚、泰国等的研究也报道了药用植物发生黄曲霉毒素污染的情况。在尼日利亚南部，研究人员发现当地常用的原始草药制剂被黄曲霉毒素污染，污染水平为0.004~0.345 $\mu g \cdot kg^{-1}$[31]。泰国28种药用植物中有5种被检测出黄曲霉毒素（1.7~14.3 $ng \cdot g^{-1}$）[32]。在印尼和马来西亚的传统草药中也检出了低水平的黄曲霉毒素[33]。

韩国的700株药用植物中，黄曲霉毒素B_1（73.27 μg·kg^{-1}）和总黄曲霉毒素（108.42 μg·kg^{-1}）阳性率分别为8.29%和2.43%[34]。在阿曼，研究人员对不同的香料进行了黄曲霉毒素和真菌菌群分析，发现优势菌群为黄曲霉、青霉菌、黑曲霉和根霉菌，其中45%的黄曲霉菌可产生黄曲霉毒素。在分析的7种香草料中，小茴香被污染的程度最高，而丁香被污染的程度最低[35]。在沙特阿拉伯的50个肉桂样品中，62%的样品被检测出黄曲霉毒素（4.67 μg·kg^{-1}）[36]。然而，在意大利，药用植物、芳香草和草药溶液样的黄曲霉毒素检测均为阴性[37]。同样，对波兰的500种草本植物样本进行黄曲霉毒素分析，认定它们均是安全的[38]。采用高效液相色谱法对印度番泻叶进行了黄曲霉毒素污染分析，结果显示仅在豆荚中检测到黄曲霉毒素，而花和叶均未受到污染[39]。ABEYWICKRAMA 等[40]在亚洲药用植物中检测到高达500 μg·kg^{-1}的黄曲霉毒素B_1。D′OUIDIO 等[41]发现人参根样品中含有15.1~16.0 ng·g^{-1}的黄曲霉毒素[41]。从水飞蓟中草药粉中分离得到的寄生曲霉菌株能产生大量的黄曲霉毒素，83个水飞蓟样品中，有19%的样品被检测出含有黄曲霉毒素（0.04~2.0 μg·kg^{-1}）[42]。

2 药用植物中的赭曲霉毒素污染

YANG 等[43]对57种中草药进行检测时发现有44%的样品污染了赭曲霉毒素A，其中92%的阳性样本明显发霉。HAN 等[44]对51个传统中草药植物样品中的赭曲霉毒素A和B同时进行分析，发现只有4个样品中含有低浓度的赭曲霉毒素。用于制备印度草药疗法阿育吠陀的草药被发现含有高达2.34 μg·g^{-1}的赭曲霉毒素A[45]。在药用植物洋椴中也检测到微量的赭曲霉毒素[46]。TRUCKSESS 等[47]对甘草样品进行分析发现其中赭曲霉毒素A含量为6 ng·g^{-1}，而GORYACHEVA 等[48]报道了甘草样品中赭曲霉毒素A污染超过10 μg·kg^{-1}。ARINU 等[49]用高效液相色谱-荧光技术对甘草根及其衍生产品进行分析，发现100%的样品赭曲霉毒素A检测呈阳性，其中一些样品被高度污染（252.8 ng·g^{-1}）[49]。在德国，对从不同药店采集的甘草根和甘草糖样品采用液相色谱-串联质谱方法进行检测，赭曲霉毒素A的污染水平在0.3~216 μg·kg^{-1}之间[50]。来自德国的另一项研究也报道了甘草根中高水平的赭曲霉毒素A污染[51]。KATERERE 等[52]对115份无花果干样品进行检测，发现有47.8%的样品检测到赭曲霉毒素A，其含量在0.12~15.31 μg·kg^{-1}范围内[52]。

3 药用植物中的伏马菌素污染

在中国、土耳其、南非和德国，都已经报道了药用植物中的伏马菌素污染。我国报道的一项芦笋中伏马菌素B_1和B_2的研究显示，80%的样品污染水平达到47~714 ng·g^{-1}[53]。有研究对8个正常和明显发霉的样品（香料、芳香草、草药）进行伏马菌素B_1和B_2污染检测，正常样品的平均污染水平分别为165.9 μg·kg^{-1}和256.8 μg·kg^{-1}，发霉样品的平均污染水平分别为129 μg·kg^{-1}和1 745 μg·kg^{-1}[54]。HAN 等[55]利用超高效液相色谱-质谱联用方法开发了一种同时检测中草药（35个样品）中伏马菌素B_1、B_2和B_3的方法，结果显示50%以上的样品被伏马菌素污染（0.58~88.95 μg·kg^{-1}），FB_1、FB_2和FB_3污染的阳性样本分别占总样本的94.4%、77.8%和33.3%；并发现在种子、根茎、草和树叶、花4类样品中，种子受污染最严重（90%），表明高含油量可能有利于伏马菌素的产生。在一项类似的研究中，研究人员也同样建立了测定药用植物中伏马菌素B_1和B_2含量的方法，样品经高效液相色谱-串联质谱检测发现有32.3%的样品总伏马菌素呈阳性[56]。

对土耳其的草药茶和药用植物进行伏马菌素分析发现，115份样品中有2份样品伏马菌素B_1呈阳性，分别为0.160 μg·g^{-1}和1.487 μg·g^{-1}[57]。土耳其的另一项研究报道了无花果干中的伏马菌素B_1污染，研究分析的115份样品中有75%被污染，平均污染水平为0.315 μg·g^{-1}[58]。在南非，30种药用野生植物中有4种被检测出含有伏马菌素B_1（8~1 553 μg·kg）[59]。对在南非销售的16个非洲传统草药样品进行检测后发现，有13个样品中含有伏马菌素B_1，含量在14~139 μg·kg^{-1}之间[60]。MARTINS 等[61]报道了柑橘树叶片中伏马菌素B_1的高度污染，污染范围在350~700 μg·kg^{-1}之间；菩提树的花叶中也发现了含量为20~200 μg·kg^{-1}的伏马霉素B_1；而洋甘菊（20~70 μg·kg^{-1}）和玉米丝样品（50~150 μg·kg^{-1}）受到的污染较轻。德国也报道了一项关于芦笋中的伏马菌素B_1和B_2的研究，发现90%的样品污染水平在36.4~4 513.7 ng·g^{-1}之间[62]。

4 药用植物中的玉米赤霉烯酮和呕吐毒素污染

我国在薏苡仁中检测到玉米赤霉烯酮的浓度范围为18.7~211.4 μg·kg^{-1}[63]。我国的另一项研究也报

道了薏苡仁中含有玉米赤霉烯酮，样品污染程度为68.9~119.6 μg·kg^{-1}[64]。YUE等[65]对58份中药材及相关产品进行了检测，结果显示只有两份薏苡仁和一份保和丸样品中存在呕吐毒素（17.2~50.5 μg·kg^{-1}）。长期以来，薏苡仁一直被用于中医治疗各种疾病，其提取物在我国也常被用于治疗癌症[66]。来自印度的一份报告显示，在130份具有重要药用价值的干根茎和块茎样品中，13.07%和6.92%的样品中存在玉米赤霉烯酮和呕吐毒素[67]。

5 药用植物中的多重真菌毒素污染

在西班牙，研究人员用ELISA法分析了药用植物和芳香类中草药的多重真菌毒素污染（黄曲霉毒素、赭曲霉毒素、玉米赤霉烯酮、伏马菌素、呕吐毒素、T-2毒素和桔霉素），结果显示84个样品中100%显示多重污染。鼠尾草和洋甘菊样品被7种真菌毒素多重污染，鼠尾草中黄曲霉毒素和赭曲霉毒素的污染范围分别为23.8~25.2 μg·kg^{-1}和1.1~17.3 μg·kg^{-1}，鼠尾草叶、洋甘菊、缬草、番泻叶和大黄的污染程度最高[68]。在土耳其的无花果干[69]和中草药（麦冬、人参、桔梗科）[70]中也检测到多重毒素（伏马菌素B_2、HT-2毒素、玉米赤霉烯酮、棒曲霉素等）污染。TAN等[71]对传统中药进行评估，发现在138个样品中仅有1个样品含有T-2毒素，T-2毒素浓度为64 ng·g^{-1}。ADLOUNI等[72]建立了HPLC检测方法，从10个黑橄榄样品中同时提取出赭曲霉毒素A、黄曲霉毒素B_1和桔霉素，定量结果显示，分别有10个和8个样品被这3种真菌毒素污染。然而在日本，经薄层色谱分析的49份药用植物样品中黄曲霉毒素、柄曲霉素和赭曲霉毒素A均为阴性，但粉末状药用植物中经常分离出曲霉菌和青霉菌[73]。

在印度，对制药工业中的草药样品进行分析，以确定真菌毒素的自然发生情况。在150份样本中，43%的样品黄曲霉毒素B_1呈阳性，6%的样品赭曲霉毒素A呈阳性，6%的样品桔霉素呈阳性，4%的样品玉米赤霉烯酮呈阳性，各毒素浓度分别为0.05~0.91、0~0.13、0~0.16 μg·g^{-1}和0~0.11 μg·g^{-1}[74]。在印度重要的药用植物（甘油三酯、大头菜、诃子）中检测到黄曲霉毒素、柄曲霉素和桔霉素[75]。然而，来自印度的肉桂、黄芥菜、印度芥菜和丁香样品没有发现多重真菌毒素污染（黄曲霉毒素、赭曲霉毒素A、柄曲霉素、桔霉素、红曲霉毒素和玉米赤霉烯酮）[76]。在巴基斯坦，约30%的药用植物样品被发现受到黄曲霉毒素和赭曲霉毒素A的污染，其中，罂粟、甘草、凝血草污染程度最高。

AZIZ等[77]报道了药用植物样品中存在黄曲霉毒素（10~160 μg·kg^{-1}）和赭曲霉毒素A（20~80 μg·kg^{-1}），在分离出的霉菌中，寄生曲霉、黄曲霉和稻曲霉可产生黄曲霉毒素，而赭曲霉和青霉菌可产生赭曲霉毒素A。在土耳其，对分别来自当地市场和出口用途的两种无花果干样品进行了黄曲霉毒素B_1和赭曲霉毒素A的污染分析，发现：在当地市场的20份样品中，只有1份样品被赭曲霉毒素A污染（2 ng·g^{-1}），所有样品均未受到黄曲霉毒素B_1污染；在拟出口的样本中，分别有23%和6%的样品黄曲霉毒素B_1（35.1 ng·g^{-1}）和赭曲霉毒素A（26.3 ng·g^{-1}）呈阳性，此外，4%的样本显示黄曲霉毒素B_1和赭曲霉毒素A共存[78]。在对生姜产品的分析中，分别在67%和74%的样品中检测到黄曲霉毒素（1~31 ng·g^{-1}）和赭曲霉毒素（1~10 ng·g^{-1}）。此外，在分析的10个人参成品中，3个被黄曲霉毒素污染（0.1 ng·g^{-1}），4个被赭曲霉毒素污染（0.4~1.8 ng·g^{-1}）[79]。

GAUTAM等[80]报道了粉状草本样品的真菌污染和真菌毒素，其中近91%的样品被真菌污染，主要为曲霉菌和青霉菌，但只有20.58%的样品被真菌毒素（黄曲霉毒素、柄曲霉素、桔霉素）污染。RIZZO等[81]研究了作为药物原料的芳香类药用植物中的产毒真菌，其中曲霉菌污染率为52%，镰刀菌污染率为27%，采用薄层色谱和高效液相色谱法检测出其中50%的菌株为产毒菌株。在另一项类似的研究中，从药用植物中分离出的真菌有89.90%属于青霉菌属和曲霉菌属，其中21.97%为产毒真菌，主要产生黄曲霉毒素和赭曲霉毒素[82]。

6 药用植物对真菌毒素的抑制作用

在世界范围内，除了关于药用植物被真菌毒素污染的相关研究，也有一些关于药用植物及其成分解毒的报道，多种药用植物精油被报道为真菌毒素和产毒真菌的抑制剂。SINGH等[83]推荐使用香樟油来抑制药用植物原料中的真菌生长和黄曲霉毒素B_1的产生。夏季香薄荷[84]和藿香蓟[85]的精油被证实对寄生曲霉菌生长和黄曲霉毒素产生具有抑制作用。同样，黄曲霉的生长和黄曲霉毒素的产生也受到小叶藤黄及其提取物[86]、印楝油[87]和香橼油[88]的抑制；柑橘提取物可降低伏马菌素和黄曲霉毒素B_1的产量[89]。

一些在动物身上进行的研究揭示了药用植物对

黄曲霉毒素毒性的保护作用。据报道，多种药用植物均可显著降低黄曲霉毒素B_1对小鼠的肝毒性，如紫菜[90]、大蒜[91]、五味子[92]、血藤[93]等。NWANKWO等[94]通过对人肝源HepG2细胞的实验，检测到一种从藤黄种子中提取到的黄酮类物质对人体中的黄曲霉毒素B_1毒性基因具有潜在抑制作用。关于药用植物提取物抗癌作用的研究中发现，银杏叶[95]和大蒜[96]提取物对黄曲霉毒素B_1诱导的肝癌发生存在有效的抑制作用。

7 真菌毒素的防控措施

开发抗性药用植物品种、使用生物和化学药剂、田间管理和收获后管理（贮藏过程中的水分控制）对预防药用植物霉菌生长和真菌毒素形成具有重要作用。关于使用生物防治剂来防止药用植物收获前真菌毒素污染的研究已多次被报道，显然，将开发抗性品种和使用具有生物竞争性的非产毒菌株的技术相结合，可能是预防药用植物真菌毒素污染最有效的策略之一。已知许多天然植物提取物和化学化合物（杀菌剂、除草剂和表面活性剂）可以防止药用植物上产毒真菌的生长和真菌毒素的形成[97]。除此之外，一些具有抗真菌特性的乳酸菌等拮抗微生物，在预防药用植物真菌毒素的形成方面也非常有效，其抗真菌特性可能涉及微生物间的竞争、热稳定性和低分子量的胞外代谢产物作用[98]。

研究人员还开发出各种物理和化学策略来防止真菌毒素污染，包括物理分离、吸附和吸附剂萃取等[99]。通过紫外光检测，对玉米、棉籽和无花果进行荧光分选，被认为是便宜且容易接受的筛选黄曲霉毒素的方法。很明显，目前没有一种物理或化学解毒方法适用于所有植物，一种真菌毒素解毒方法的有效性取决于水分含量、温度以及真菌毒素的类型、浓度和真菌毒素与成分之间的结合程度等。一系列的化学化合物，包括盐酸、氨、过氧化氢、臭氧、亚硫酸氢钠等，发现其中氯在真菌毒素解毒方面似乎具有很好的潜力，但使用氯会显著降低食品的营养价值，或在处理过的产品中产生具有不良感官特性的有毒衍生物，这将严重限制氯的广泛使用。因此利用生物防治剂在控制和减少药用植物真菌毒素污染方面具有巨大潜力，其也被广泛认为是替代化学杀菌剂的良好材料。

8 结论与展望

药用植物真菌毒素污染是一种普遍的全球性事件。很明显，各类药用植物和相关产品都可能被真菌毒素污染，并超过国际上一些国家规定的阈值限制。因此，对药用植物中真菌毒素污染的监测十分必要。用于医疗用途的植物材料在使用前应妥善储藏并评估是否存在真菌毒素污染，以确保其安全性。消费者所食用的药用植物种类和数量随其健康状况的不同而不同，因此需要对每一种类型的药用植物单独考虑人体因摄入药用植物而暴露于真菌毒素的浓度水平。此外，各种研究报告显示，从药用植物材料中分离出来的真菌具有产毒潜力。因此，为了降低真菌毒素对人体健康的危害，应该从多个方面做好产毒真菌和真菌毒素污染的防控工作。

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