on Adulteration of *Hydrastis canadensis* root and rhizome

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Goal: The goal of this bulletin is to provide information and/or updates on issues regarding adulteration of goldenseal (*Hydrastis canadensis*) root to the international herbal industry and extended natural products community in general. It is intended to present the available data on occurrence of adulteration, the market situation, and consequences for the consumer and the industry.

1 General Information

1.1 Common name: Goldenseal

1.2 Other common names:

*English*: Yellow root, yellow puccoon, ground raspberry, wild curcuma, Indian dye, eye root, eye balm, Indian paint, jaundice root, Warnera

*French*: Hydraste du Canada, hydraste, fard inolien, framboise de terre, sceau d’or

*German*: Goldsiegelwurzel, Kanadische Gelbwurz, Kanadische Orangenwurzel

*Italian*: Idraste, radice gialla

*Spanish*: Hidrastis, hidrastis de Canadá, raíz de oro, scello de oro

1.3 Accepted Latin binomial: *Hydrastis canadensis*

1.4 Botanical family: Ranunculaceae

1.5 Plant part and form: Whole fresh or dry roots and rhizomes, powdered dry roots and rhizomes, hydroalcoholic and glycerin-water extracts and powdered dry extracts. Dried whole or powdered roots and rhizomes complying with the *United States Pharmacopeia* (USP) are required to contain not less than 2.0% of hydrastine and not less than 2.5% berberine.  

Goldenseal *Hydrastis canadensis*  
Photo ©2016 Steven Foster
1.6 General use(s): Native American tribes used goldenseal root and rhizome preparations as eye washes, treatments for skin disorders, bitter tonics, and for respiratory ailments and the infectious diseases brought by European settlers.11 The plant was included in The American Eclectic Materia Medica and Therapeutics12 and King’s American Dispensatory,13 which increased its use substantially among Eclectic physicians for infections, mouth ulcers and thrush, inflamed mucous membranes, chronic gonorrhea, jaundice, gastrointestinal complaints, as a bitter tonic and as a uterine tonic. Goldenseal preparations are now used as antimicrobial agents to treat infections of the mucosal membrane, including mouth, upper respiratory tract, gastrointestinal tract, eyes, vagina, as well as for wounds.9

2 Market

2.1 Importance in the trade: Echinacea-goldenseal combination dietary supplements were ranked #15 in sales in the natural food channel in the United States in 2013 and in 2014 (Table 1), with estimated sales exceeding $5 million in 2014. Sales in the Mainstream Multi-Outlet retail channel (excluding sales data from Walmart and Club stores in 2013 and 2014, which were not available) were lower, with echinacea-goldenseal products ranking between #41 and #53. (T. Smith [American Botanical Council] e-mail to S. Gafner, September 2, 2015 and September 3, 2015)14,15

Goldenseal root/rhizome-only supplements did not rank in the top 50 best-selling herbal supplements in the natural food channel or the Mainstream Multi-Outlet retail channel from 2012-2014 (Table 2).

2.2 Supply sources: Historically, the majority of the goldenseal root and rhizome on the market has come from wild-harvested material, with Kentucky and Tennessee as the major producers. Other states along the Appalachian Mountains and in the northeastern United States provide additional supplies.9 There is no credible evidence that H. canadensis is cultivated commercially outside of the United States and Canada.

2.3 Conservation status: In 1997 goldenseal was listed on Appendix II of the Convention on International Trade in Endangered Species of Wild Fauna and Flora,17 which also controls global trade and markets for H. canadensis.18 Several states consider H. canadensis in need of conservation: Pennsylvania lists the plant as vulnerable, at high risk of endangerment in the wild; Maryland, Michigan and New York list the plant as threatened, at risk of extinction in the wild sometime in the near future; Connecticut, Georgia, Massachusetts, Minnesota, New Jersey and Vermont list the plant as endangered, at high risk of extinction in the wild; North Carolina and Tennessee list the plant as endangered with special concern, at critically high risk of extinction in the wild.19

2.4 Raw material forms: Bulk goldenseal root and rhizome raw material is sold as whole roots and rhizomes (fresh or dry), cut and sifted, or as powder.

2.5 Market dynamics: Despite a general trend of price increases since 1986, the costs vary considerably from year to year. Prices per kg paid to collectors of dried wild goldenseal root and rhizome ranged from US $44–$77 between 1996 and 2005, and $77 wild to $110 for organic woods-cultivated material for the same time frame.20 The price per kg of dried wild root and rhizome peaked at $77 in 2001, dropping to $44 per kg in 2005, and fluctuating between $44–55 per kg through 2010.21 In its tonnage report, the American Herbal Products Association (AHPA) reported an average yearly harvest of 10 metric tons (MT) of cultivated compared to 30 MT of wild-harvested dry goldenseal root and rhizome between 2004 and 2010. The amounts of fresh goldenseal root and rhizome have remained below one MT in the same timeframe. Generally, the quantities of goldenseal root and rhizome harvested between 2004 and 2010 have been relatively steady.22

3 Adulteration

3.1 Historical adulterants: At the height of the Eclectic medical movement in the United States during the early 1900s, the price of goldenseal had risen to the point that

Table 1: Sales data for echinacea-goldenseal dietary supplements from 2012-2014.

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Table 2: Sales data for goldenseal-only dietary supplements from 2012-2014.

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<tr>
<td>Natural</td>
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aAccording to SPINS (SPINS does not track Whole Foods Market sales, which is a major natural products retailer in the US)
bAccording to SPINS/IRI (the Mainstream Multi-Outlet channel was formerly known as food, drug and mass market channel [FDM]; possible sales at Walmart and Club stores are excluded in 2013 and 2014)
n/a: not available

Source: T. Smith (American Botanical Council) e-mail September 2, 2015 and September 3, 2015
several plant species were being used as economic adulterants on a regular basis; these included goldthread or coptis (Coptis spp., Ranunculaceae), yellow root (Xanthorrhiza simplicissima, Ranunculaceae), European peony (Paeonia officinalis, Paeoniaceae), and twin leaf (Jeffersonia diphylla, Berberidaceae). The adulteration of goldenseal with Virginia snakeroot (Aristolochia serpentaria, Aristolochiaceae), which is of concern due to its content of nephrotoxic and carcinogenic aristolochic acids, was initially documented in 1892, and described again in 1900. However, there are no reported cases of adulteration with Virginia snakeroot in recent times. Additional historical adulterants and contaminants of goldenseal that have been documented include Athyrium filix-femina (Athyriaceae), Stylophorum diphyllum (Papaveraceae), Cypripedium calceolus (Orchidaceae), Collinsonia canadensis (Lamiaceae), Trillium spp. (Melanthiaceae), Caulophyllum thalictroides (Berberidaceae), and Polygala senega (Polygalaceae) as admixtures or contaminants. It is unlikely that adulteration is occurring with these species in commerce today.

3.2 Recent adulterants: Over the past 20–30 years, economic adulterants have reappeared in goldenseal products, based in part on erroneous use of goldenseal to negate illicit drug testing in the 1980s. The adulterating species include Japanese goldthread (Coptis japonica), yellow root (Xanthorrhiza simplicissima), Oregon grape (Mahonia aquifolium, Berberidaceae), celandine (Chelidonium majus, Papaveraceae), barberry (Berberis spp., Berberidaceae), and yellow dock (Rumex spp., Polygonaceae) root. There is a single case of the sale of other root materials labeled as goldenseal, e.g., plantain (Plantago spp., Plantaginaceae) root, nettles (Urtica dioica, Urticaceae) root, or passionflower (Passiflora incarnata, Passifloraceae) root colored with a yellow dye, as reported on the website of an analytical laboratory.

Historically, market pricing for H. canadensis has displayed instability. As noted above, over the more recent 15 years the price has ranged between $44-110/kg for dry root and rhizome material. Assuming that the relatively higher goldenseal price level drives intentional economic adulteration, the addition and/or substitution with several of the adulterating plant species would represent substantial cost savings to an unscrupulous supplier. Over the past 20 years, dried roots of the species Mahonia have sold at $6.1-$8.8 per kg and Berberis at $7.3-14.30 per kg. Over the past 10 years the price/kg of adulterating species ranged as follows: celandine $3.7-6.9, barberry $5.6-19.3, yellow dock imported $9.5-10.1 and domestically grown $12.9-14.8. The price/kg for Coptis spp. was approximately $14.1 per kg in 2011 and $11.0/kg in 2015.

3.3 Sources of information supporting confirmation of adulteration: Goldenseal adulteration can be detected through the presence or absence of several alkaloids, namely the presence of berberine, canadine, hydrastine, and hydastinine, and the absence of palmatine. Although most species used as adulterants contain berberine, the alkaloids hydrastine and canadine are unique to goldenseal. The presence of palmatine is indicative of adulteration by Coptis spp., the most common adulterant of goldenseal. An analysis of several dietary supplement products marketed as goldenseal root extract using the AOAC official method 2008.04-2008, a high-performance liquid chromatography (HPLC) method for the analysis of goldenseal material, revealed that several products contained palmatine.

A validated HPLC-mass spectrometry (MS) method was used to analyze H. canadensis root from three suppliers along with the common adulterants – Coptis spp. root, M. aquifolium root, Berberis spp. bark, and C. majus herb. Of the three commercial lots that were purchased, all contained the expected goldenseal alkaloids: hydastinine berberastine, tetrahydroberberastine, canadine, berberine, hydrastine, and canadine. However, one product contained additional alkaloids not associated with goldenseal – palmatine, coptisine, and jatrorrhizine – thus suggesting admixture of an adulterating species. Avula et al. used an ultra-high performance liquid chromatography (UHPLC) method to detect the non-goldenseal constituents palmatine, coptisine and jatrorrhizine in a commercial goldenseal product. An emerging adulterant problem stems from the use of goldenseal leaf material, which contains both berberine and hydastine, but in a different ratio from goldenseal root.

A recent report by a company specializing in DNA-based species identification analyzed several off-the-shelf goldenseal products, including those from a company described as “a major manufacturer.” The company used their proprietary ConfirmIDNATM method, a DNA barcoding method using universal primers, to identify plantain root, nettle root, or passionflower root rather than goldenseal. In addition, the product was colored with a yellow dye. No additional tests were carried out to confirm these findings.

Although this publication is focused on H. canadensis root, commercial trade in H. canadensis leaf does occur. Dried leaf harvest for the years 2004-2010 was estimated to range from 3.5-8.5 MT. During that time the price was approximately $2.2-11.0/kg (E. Burkhart [Pennsylvania State University] e-mail, September 21, 2012). Goldenseal leaf is an article of commerce, and there are allegations of its use as a low-cost adulterant. Actual evidence of non-declared goldenseal leaf as an adulterant to goldenseal root/rhizome is rare. One analysis of hydrastine and berberine in goldenseal leaf found levels of these alkaloids ranging at 0.27-0.29% and 0.36-0.39%, respectively, while levels in the root were 2.25-3.32% and 2.61-3.75%, respectively. As part of this 2002 study, three commercial echinacea/goldenseal products were tested with one containing only berberine in the expected range of goldenseal-derived isoquinoline alkaloids, which may indicate an adulterant species was used. Another contained very low alkaloid content suggesting possible leaf adulteration.

3.4 Accidental or intentional adulteration: Historically as well as recently, the use of adulterating species appears to be motivated primarily by economic gain, particularly when bulk goldenseal root is selling for up to $110 per kg.
3.5 Frequency of occurrence: There is no comprehensive study on the frequency of goldenseal adulteration. One analysis in 2003 of three lots of purchased commercial goldenseal root powder found goldenseal alkaloids hydastinone, berberastinone, tetrahydroberberastine, canadoline, berberine, hydastinone, and canadine in all samples, while only one sample from a single supplier also contained palmatine, coptisine, and jatrorrhizine, presumably indicating that adulteration with coptis occurred in that one sample.39

3.6 Possible therapeutic/safety issues: Although no systematic investigation into human toxic effects associated with the use of Berberis spp., Coptis spp., or M. aquifolium could be found, no other spontaneous or anecdotal reports of adverse effects could be found. The second edition of the American Herbal Products Association’s Botanical Safety Handbook (BSH2) lists B. vulgaris and C. chinensis as class 2b safety ingredients, meaning that these botanicals should not be used during pregnancy. The safety concerns in the BSH2 are based on studies using pure berberine, and may not directly apply to extracts made from barberry or coptis. Mahonia aquifolium, which also contains berberine, is presented as a safety class 1 ingredient, which is a botanical that is considered to be safe when used appropriately. Nevertheless, use of Oregon grape during pregnancy is not recommended. In addition, all berberine-containing plants are not recommended for use during lactation.35

Case studies of Chelidonium majus herb in Germany have associated consumption of the herb with liver toxicity. Ad hoc causality assessments in 22 spontaneous cases employing a liver-specific, standardized, quantitative assessment method (Council for International Organizations of Medical Sciences) found causality to be highly probable (n = 2), probable (n = 6), possible (n = 10), unlikely (n = 1), and excluded (n = 3). The pattern of liver injury was observed predominantly among female consumers. The average treatment was 36.4 days, and the latency period until first symptoms and jaundice was 29.8 and 35.6 days, respectively. The study did not identify which of the constituents were responsible for the liver injury.36 No product analysis was conducted and manufacturer observance of cGMP (current Good Manufacturing Practices) was assumed. As such, it is unknown if adulterated products impacted the reported adverse effects.

3.7 Analytical methods to detect adulteration: Brown and Roman conducted a multi-laboratory collaborative study utilizing a HPLC-ultraviolet (UV) detection method, previously validated using AOAC International single-laboratory validation guidelines, to measure hydastinone and berberine in goldenseal root raw materials, extracts, and dietary supplements at concentrations of 0.4 to 6% (w/w). In addition to the quantification of berberine and hydastinone, the method also detected the presence of palmatine, an indicator of adulteration with Coptis spp.35 Based on the results of the study the method was subsequently adopted as AOAC official method 2008.04-2008.37 Weber et al., generated different alkaloid profiles for H. canadensis root and two berberine-containing Coptis species using a validated HPLC-MS method.38 Kamath and colleagues investigated alkaloid compositions of H. canadensis, American goldthread (C. trifolia) and coptis (C. chinensis). They determined that the spectrum of alkaloids in C. chinensis was different from those in H. canadensis and C. trifolia, showing that the alkaloid fingerprint was suitable to distinguish the species.39 Another HPLC-MS method was shown to separate 10 analytes (berbamine, berberine, canadine, chelerythrine, coptisine, hydastinone, hydastidine, jatrorrhizine, palmatine and sanguinarine) from six different plant species (H. canadensis, Coptis japonica, B. vulgaris, Chelidonium majus, M. aquifolium and S. canadensis), allowing analysts to quantify the presence of adulterants at concentrations as low as 5%.40 More recently, a UPLC method with UV detection was used to identify the non-goldenseal constituents palmatine, coptisine, and jatrorrhizine in a commercial goldenseal product.34

Govindan and Govindan developed a thin-layer chromatography (TLC) method to detect hydastinone, hydastidine, and berberine of several goldenseal preparations.41 Their analysis identified three samples containing only berberine, and one sample that contained none of the alkaloids, potentially indicating economic adulteration. The results of the TLC analysis were confirmed by subsequent HPLC tests.41 The American Herbal Pharmacopoeia adopted a validated high performance TLC (HPTLC) method that simultaneously detects palmatine, which is found in adulterants, plus berberine, hydastidine, and hydastinone. Upton noted the usefulness of hydastinone as a reliable marker for old or poor quality H. canadensis, since it is formed as a degradation product of hydastidine.42 Finally, a TLC/desorption electrospray ionization (DESI)-MS method was shown to detect non-goldenseal alkaloids from adulterants in goldenseal products.43

Several official compendial methods exist that may also be applied to adulterant detection, including the European Pharmacopoeia44 and the United States Pharmacopeia–National Formulary.45 Criteria to perform identification using macroscopic and microscopic examinations of goldenseal rhizome and root are presented in several references.32,44,45 In addition to the macroscopic and microscopic characteristics of goldenseal, the AHP monograph also lists such characteristics for Oregon grape, Coptis spp., yellow dock, and yellow root.42

Other, unique analytical systems have been developed that detect both major and minor H. canadensis alkaloids. These methods, including capillary electrophoresis-mass spectrometry (CE-MS),46 pH-zone refining counter current chromatography (CCC),47 shift subtracted Raman spectroscopy (SSRS),48 and an enzyme-linked immunosorbent assay (ELISA) linked to a HPLC,49 may be adapted for the detection of adulterants.

4 Conclusions

Habitat destruction in some areas of the eastern United States, goldenseal’s native range, has decreased the availability of this important medicinal plant. Pricing pressure has
historically increased the incentive for economically motivated adulteration of goldenseal root. Although the actual extent of adulteration of goldenseal root and rhizome in the current market is not clear, a number of authenticated methods exist to detect such adulteration, including some that have been validated.

*Although it has sometimes been called wild curcuma, goldenseal should not be confused with turmeric root (Curcuma longa, Zingiberaceae).

5 References


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**REVISION SUMMARY**

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<th>Date Revised</th>
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