Differential Contribution of Antioxidants to Antioxidative Functions in Galls Evaluated by Grey System Theory

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Abstract — The contents of five antioxidants and the abilities of four antioxidative functions were determined in eight insect galls and their six host plants leaves. The grey relational order indicated that scavenging DPPH free radicals ability was strongly related to polyphenols and carotenoids, chelating ferrous ions activity strongly related to anthocyanins and polyphenols, superoxide anion scavenging activity related to polyphenols and flavonoids, and reducing power related to carotenoids and polyphenols. The preference order evaluated by grey decision making for antioxidative capacity was *Schlechtendalia Chinensis* (gall) > *Daphnephila sueyenae* (gall) > Unknown sp. 2 (gall) > *Daphnephila taiwanensis* (Gall) > *Bruggmanniella* sp. (gall) > *Ceratovacuna nekoashi* (gall) > *Styrax suberifolia* (leaf) > *Sycopsis sinensis* (leaf) > *Styrax formosana* (leaf) > Unknown sp. 1 (gall) > *Litsea acuminata* (leaf) > *Machilus thunbergii* (leaf) > *Pseudoregma bambucicola* (gall) > *Ficus erecta* var. beechyana (leaf). The contents of polyphenols were the most relevant to the antioxidant ability, and that of chlorophylls is the less. The galls had higher antioxidative capacity than their host leaves to scavenge free radicals effectively for balancing the increases of free radicals in their bodies.

Keywords: Gall, Host plant; Antioxidant; Antioxidative capacity; Grey decision making; Grey relational analysis.

Introduction

Galls are derived from plant tissues induced by insects. Plant secondary metabolites played important roles in the defense against free radicals. The levels of secondary metabolites, such as anthocyanin and tannin and phenolic compounds in the gall or galled foliar tissue were siginificantly higher than that in the non-galled foliar tissues [1,2,3].

Unbalance between free radicals generation and antioxidant defense led to oxidative stress and cellular injury. Probably, the process of galls formation causing oxidative damage of plant cells was similar to physical injury. Mittapalli *et al.* revealing that increased antioxidant activity from the galling insects was due to the resistance of plant cells generating a large number of free radicals in the process of oxidative damages [4]. It was reported that the antioxidant activity in *Quercus infectoria* galls pose potent antioxidative capacity. High capacity of free radical scavenging and high quantity of ethanol extract of gall may suppress oxidative damages [5]. In addition, the nutgall, i.e. Compendium of *Materia Medica* made by aphid had been used as a traditional Chinese medicine herb due to high level of antioxidative capacity [6].

The grey system theory was initially presented by Deng in 1982 [7,8]. Grey relation analysis has been successfully used to compare the contribution of individual antioxidants to total antioxidative capacity or function in various measurement systems [9 and its references]. The grey decision making was used to identify which herb plants had the highest antioxidative capacity and function for marketing [10 and its references].

In this study, grey relational analysis was hired to pinpoint the most important contribution or to compare the contribution degrees of the individual antioxidants to the relative antioxidative functions in the gall tissues and the leaves of their host plants. In addition, grey decision making was further used to evaluate the antioxidative capacity and the optimum alternative in the gall tissues and the leaves of host plants.

Materials and methods

Insect galls and host plants

Eight kinds of common gall were collected from different parts of Taiwan.. Among them, five galls are located on leaf, one on stem and two on buds (Table 1).

Table 1. The gall-inducing insects and their host plants.

Galls	Gall-inducing insects	Host plants	Location
Oval-pointed galls	Diptera: Cecidomyiidae *Daphnephila taiwanensis*	Lauraceae *Machilus thunbergii*	Leaf
Obovate gall	Diptera: Cecidomyiidae *Daphnephila sueyenae*	Lauraceae *Machilus thunbergii*	Leaf
Cup galls	Diptera: Cecidomyiidae *Bruggmanniella* sp.	Lauraceae *Litsea acuminata*	Leaf
Rolling-sphere galls	Diptera: Cecidomyiidae Unidentified sp. 1	Moraceae *Ficus erecta* var. beechyana	Leaf
Green gall	Hemiptera: Hormaphididae Unidentified sp. 2	Hamamelidaceae *Sycopsis sinensis*	Stem
Cat's claw-shaped galls	Hemiptera: Hormaphididae *Pseudoregma bambucicola*	Styracaceae *Styrax suberifolia*	Bud
Cat's claw-shaped galls	Hemiptera: Hormaphididae *Ceratovacuna nekoashi*	Styracaceae *Styrax formosana*	Bud
Nutgalls	Hemiptera: Pemphigidae *Schlechtendalia chinensis*	Anacardiaceae *Rhus javanica* var. roxbur-ghiana	Leaf

Determination of antioxidants contents and antioxidative functions

The galls samples and the disinfected-leaves of host plants were chopped separately, frozen in liquid nitrogen, and ground to a fine powder. The contents of antioxidants, such as polyphenols, flavonoids, anthocyanins, chlorophylls and carotenoids were determined. The antioxidative functions, including 1, 1-diphenyl-2-picrylhydrazyl (DPPH) free radical scavenging ability, chelating activity of ferrous ions (Fe^{2+}), scavenging activity of the extracts towards superoxide anion and reducing power were further measured [10 and its references]

Grey relational analysis

The contents of five antioxidants were used as a comparison or test series, and antioxidative functions, DPPH scavenging activity, Fe^{2+} ion scavenging activity, superoxide anion scavenging activity and reducing power, were used as a reference series, respectively. The basic principle and method of grey relational analysis were as follows [7,8].

(1) Normalization of original data series

If the original data was $x_0=(x(1), x(2), \cdots x(n))$, then the initial normalization data series was $x_0^{*}= \left(\frac{x(1)}{x(1)}, \frac{x(2)}{x(1)}, \cdots, \frac{x(n)}{x(1)}\right)$. If the mean value of the original data was $\overline{X}$, then the mean value normalization data series became $x_0^* = \left(\frac{x(1)}{\overline{X}}, \frac{x(2)}{\overline{X}}, \cdots, \frac{x(n)}{\overline{X}}\right)$ where $\overline{X} = \frac{1}{n}\sum_{k=1}^{n} x(k)$.

(2) The grey relational coefficient γ was defined as follows:

$$\gamma(x_0(k), x_j(k)) = \frac{\min_j \min_k |x_0(k) - x_j)k)| + \zeta \max_j \max_k |x_0(k) - x_j)k)|}{|x_0(k) - x_j)k)| + \zeta \max_j \max_k |x_0(k) - x_j)k)|}$$

where j=1 ,2, $\cdots$, m; k =1,2, $\cdots$, n, ξ was the identification coefficient; its value was taken as [0,1], usually, $\xi = 0.5$. x_0 was the reference data series. x_j was the test data series. $|x_0(k) - x_j(k)|$ was the absolute value (norm) of the difference between $x_0(k)$ and $x_j(k)$. $\min_j \min_k |x_0(k) - x_j(k)|$ was called the secondary minimum difference which was selected in all j. $\min_k |x_0(k) - x_j(k)|$ was called the first minimum difference which was selected in all k. All the same as *max max*.

(3) The grey relational grade $\gamma(x_0, x_j)$ was:

$$\gamma(x_0, x_j) = \frac{1}{n}\sum_{k=1}^{n} \gamma(x_0(k), x_j(k)).$$

(4) Rearrangement of grey relational grade from large value to little value in a series The grey relational grade had the following characteristics:

a. $0 \le \gamma(x_0, x_j) \le 1$.

b. The more similar the test data $x_j(k)$ was to the reference data $x_0(k)$, the larger the grey relational grade value γ.

c. Only if the reference series $x_0(k)$ was exactly equal to the test series $x_j(k)$, did the grey relational grade value γ^*=1.

The model of grey decision making

When it came up event (A), strategy (B) was used to deal with situation (S). While choosing the best strategy to deal with the events, it's called Decision-Making. The definition and algorithm of the Grey Decision Making method were as follows [7,8]:

Let $S_{ij}=(a_i,b_j)$ stood for situation, a_i for the event by antioxidative capacity evaluation,

b_j for galls and leaves, u_{ij}^p stood for the sample of situation S_{ij} in the pth criteria.

Polarity transformation u_{ij}^p for [0,1]

$$M(u_{ij}^p) = r_{ij}^p \in r^p \Rightarrow r_{ij}^p \in [0,1]$$

Upper Effect Measure (UEM)

$$P_{OL(\max)} \rightarrow r_{ij}^p = \frac{u_{ij}^p}{\max\limits_j u_{ij}^p}$$

Lower Effect Measure (LEM)

$$P_{OL(\min)} \rightarrow r_{ij}^p = \frac{\min\limits_j u_{ij}^p}{u_{ij}^p}$$

Medium Effect Measure (MEM)

$$P_{OL(mem)} \rightarrow r_{ij}^p = \frac{\min\{u_{ij}^p, u_0^p\}}{\max\{u_{ij}^p, u_0^p\}} \quad \text{where } u_0^p = \frac{1}{m}\sum_{j=1}^{m} u_{ij}^p$$

Calculated the synthesis effect measure r_{ij}^{Σ}: $r_{ij}^{\Sigma} = \frac{1}{l}\sum_{p=1}^{l} r_{ij}^p$

To make certain optimum alternative $r_{ij^*}^{\Sigma}$: $r_{ij^*}^{\Sigma} = \max\{r_{i1}^{\Sigma}, r_{i2}^{\Sigma}, \ldots, r_{im}^{\Sigma}\}$

Results and Discussion

Polyphenols contents from the galls were significantly higher than those from their host plants, except for the unknown 2 galls on the *S. sinensis*. The highest and lowest polyphenols contents were found in the *S. chinensis* galls and the leaves of the *F. erecta*, respectively. The highest and lowest levels of flavonoids were observed in the leaves of the *F. erecta* and *S. chinensis* galls, respectively. The *D. taiwanensis* galls on the *M. thunbergii* showed significantly higher anthocyanins content than others. Compared with the host plants leaves, chlorophylls in the insect-induced galls drastically decreased by approximately 4-18 fold while carotenoids declined by about 1.6-4 fold, except for the leaves of *S. suberifolia* (Table 2).

The ability of various antioxidative functions from the methanol extract of galls and their host plant leaves was presented in Table 3.

Table 2. The contents of various antioxidants in galls and their host plants.

Galls and host plants	Polyphenols (mg gallic acid/DW g)	Flavonoids (μg/DW g)	Anthocyanins (μmol/DW g)	Chlorophyll (μg/DW g)	Carotenoids (μg/DW g)
Daphnephila taiwanensis (gall)	177.5	55.1	1.37	532	210
Daphnephila sueyenae (gall)	89.9	20.3	0.39	791	296
Machilus thunbergii (leaf)	23.8	49.3	0.44	5001	572
Bruggmanniella sp. (gall)	101.8	8.8	0.26	255	80
Litsea acuminata (leaf)	25.4	64.9	0.06	2758	342
Unidentified sp. 1 (gall)	89.7	16.0	0.52	904	482
Ficus erecta var. beechyana (leaf)	19.9	116.4	0.94	12031	763
Unidentified sp. 2 (gall)	99.2	5.6	0.09	179	118
Sycopsis sinensis (leaf)	142.7	33.0	0.14	4284	468
Pseudoregma bambucicola (gall)	126.4	12.9	0.24	700	436
Styrax suberifolia (leaf)	26.4	37.9	0.39	3001	91
Ceratovacuna nekoashi (gall)	94.7	6.5	0.26	326	378
Styrax formosana (leaf)	57.7	91.0	0.89	5782	759
Schlechtendalia chinensis (gall)	821.1	5.5	0.10	62	85

Table 3. The ability of various antioxidative functions from galls and their host plants leaves.

Galls and host plants	P1:DPPH activity (%)	P2: Fe^{2+} ion scavenging activity (%)	P3:Superoxide anion scavenging activity (%)	P4: Reducing power
b1:Daphnephila taiwanensis (gall)	87.41	40.25	47.68	2.19
b2:Daphnephila sueyenae (gall)	85.46	60.68	57.66	2.18
b3:Machilus thunbergii (leaf)	83.50	19.59	27.80	2.18
b4:Bruggmanniella sp. (gall)	89.91	20.96	46.72	2.20
b5:Litsea acuminata (leaf)	79.75	27.64	24.45	2.18
b6:Unknown sp. 1 (gall)	87.82	24.22	25.03	2.19
b7:Ficus erecta var. beechyana (leaf)	6.22	57.82	0.39	0.65
b8:Unknown sp. 2 (gall)	90.20	41.15	54.95	2.22
b9:Sycopsis sinensis (leaf)	84.97	13.61	43.50	2.23
b10:Pseudoregma bambucicola (gall)	83.10	10.35	13.19	2.22
b11:Styrax suberifolia (leaf)	85.57	27.89	43.37	2.18
b12:Ceratovacuna nekoashi (gall)	85.46	5.32	59.65	2.19
b13:Styrax formosana (leaf)	83.30	24.41	28.76	2.30
b14:Schlechtendalia chinensis (gall)	91.68	82.98	53.73	2.32

All methanol extract of galls and their host plants exhibited a relatively higher percentage of scavenging DPPH radicals than the leaves of the *F. erecta*. The highest and lowest chelating activity of ferrous ions were found in the extract of the *S. chinensis* nutgalls and the *C. nekoashi* cat's claw-shaped galls, respectively. The highest and lowest scavenging activity of the extracts towards superoxide anion showed in the *C. nekoashi* galls and the leaves of the *F. erecta*, respectively. The leaves of the *F. erecta* contained a significant lower level of reduction power compared to other extracts of the galls and the leaves of host plants.

The grey value and grey order for each antioxidant were shown in Table 4. When the antioxidants were listed in descending order of the contribution they make to the DPPH scavenging activity, the order was polyphenols> carotenoids > anthocyanins> flavonoids > chlorophyll. The grey relational value were anthocyanins> polyphenols > flavonoids > carotenoids > chlorophyll to the Fe^{2+} ion scavenging activity, polyphenols > flavonoids> anthocyanins > carotenoids > chlorophyll to superoxide anion scavenging activity, and carotenoids> polyphenols> anthocyanins> flavonoids > chlorophyll to reducing power, respectively.

Table 4. The grey relational value and order of antioxidants contents and antioxidative functions in the galls and their host plants leaves.

Antioxidative functions	Antioxidants				
	Polyphenols	Flavonoids	Anthocyanins	Chlorophyll	Carotenoids
DPPH scavenging activity	0.825 (1)	0.774 (4)	0.793 (3)	0.742 (5)	0.824 (2)
Fe^{2+} ion scavenging activity	0.749 (2)	0.739 (3)	0.754 (1)	0.686 (5)	0.700 (4)
superoxide anion scavenging activity	0.806 (1)	0.748 (2)	0.742 (3)	0.716 (5)	0.739 (4)
Reducing power	0.825 (2)	0.781 (4)	0.799 (3)	0.749 (5)	0.830 (1)

* The number in parenthesis was grey order.

The most frequently mentioned antioxidants in plants were carotenoids, flavonoids, anthocyanins, phenolics, tannins, polyphenols, and vitamins [11,12]. Chlorophyll has been found to exhibit antioxidant activity in rat liver, and also by means of its ability to protect mitochondria from oxidative damage induced by various reactive oxygen species [13]. In this study, various antioxidants made different contributions to the four antioxidative functions.

For the DPPH scavenging activity, the polyphenols and carotenoids played much more important roles than other antioxidants. Free radical scavenging was a mechanism for antioxidant to inhibit lipid oxidation [14], and scavenging of DPPH free radicals by antioxidants was due to takes place with phenolics and flavonoids [15]. The result of grey relational analysis agree with other works, which showed that scavenging of DPPH free radicals was relevant to polyphenols and carotenoids [16,17].

The anthocyanins and polyphenols played much more important roles in determining the Fe^{2+} ion scavenging activity based on grey relational analysis (Table 4). Among metal ions, Fe^{2+} was the most siginificant material to promote oxidation. The anthocyanins extracted from litchi (*Litchi chinenesis* Sonn.) fruit pericarp tissues were inhibited deoxyribose degradation induced by hydroxyl radical, mainly via chelating Fe^{2+} ion rather than scavenging hydroxyl radical directly [18]. The ethanolic extract of *Quercus infectoria* galls showed relative high chelating activity and polyphenols content. Probably its high content of polyphenols played a considerable role [19].

The grey relational order revealed that polyphenols and flavonids had a larger effect on superoxide anion scavenging activity, and polyphenols and carotenoids played much more important roles in determining reducing power. Higher amount of polyphenols and flavonoids had better ability to remove superoxide anion. Reducing power exerted antioxidant activity by donating a hydrogen atom to break the chain of free radical, inhibit the occurrence of lipid oxidation, and can be used as a provider of electronics [14]. The reducing power of green alga extract indicated that the marked antioxidant activity of algae was believed to be due to the presence of carotenoids [18]. Kaur *et al.* (2008) reported that a large amount of polyphenols in *Quercus infectoria* gall extract possessed a high reducing capacity [19]. Strong relationship between carotenoids, polyphenols and reducing power was also found in the leaves of radish [20], and in addition to carotenoids most of the antioxidative effect in plants was mainly due to the presence of phenolic compounds [21].

The antioxidative capacity evaluation had put 14 alternatives set $B=\{b_1,b_2,\cdots,b_{14}\}$. The study was then subjected to 4 criteria set $P=\{p_1,p_2,p_3,p_4\}$ as part of an evaluation of the alternatives. These included DPPH scavenging activity、Fe^{2+} ion scavenging activity、superoxide anion scavenging activity and reducing power (Table 2).

In the four antioxidative functions as criteria, all of them were of the maximum polarity. A calculation of the polarity transformation was shown in the following matrix.

$$r_{ij}^{p} = \begin{array}{c} \\ b_1 \\ b_2 \\ b_3 \\ b_4 \\ b_5 \\ b_6 \\ b_7 \\ b_8 \\ b_9 \\ b_{10} \\ b_{11} \\ b_{12} \\ b_{13} \\ b_{14} \end{array} \begin{array}{c} \begin{array}{cccc} p_1 & p_2 & p_3 & p_4 \end{array} \\ \begin{bmatrix} 0.95 & 0.49 & 0.80 & 0.94 \\ 0.93 & 0.73 & 0.97 & 0.94 \\ 0.91 & 0.24 & 0.47 & 0.94 \\ 0.98 & 0.25 & 0.78 & 0.95 \\ 0.87 & 0.33 & 0.41 & 0.94 \\ 0.96 & 0.29 & 0.42 & 0.94 \\ 0.07 & 0.70 & 0.01 & 0.28 \\ 0.98 & 0.50 & 0.92 & 0.96 \\ 0.93 & 0.16 & 0.73 & 0.96 \\ 0.91 & 0.12 & 0.22 & 0.95 \\ 0.93 & 0.34 & 0.73 & 0.94 \\ 0.93 & 0.06 & 1.00 & 0.94 \\ 0.91 & 0.29 & 0.48 & 0.99 \\ 1.00 & 1.00 & 0.90 & 1.00 \end{bmatrix} \end{array}$$

Calculated the synthesis effect measure r_{ij}^{Σ}:

$$r_{ij}^{\Sigma} = \frac{1}{4}\sum_{p=1}^{4} r_{ij}^{p}, (i=1; j=1,2\cdots,14; p=1,2,\cdots,4)$$

j=1, then $r_{11}^{\Sigma} = (0.95 + 0.49 + 0.80 + 0.94)/4 = 0.79$

Similarly, there were

$r_{12}^{\Sigma} == 0.89$, $r_{13}^{\Sigma} == 0.64$, $r_{14}^{\Sigma} == 0.74$, $r_{15}^{\Sigma} == 0.64$, $r_{16}^{\Sigma} == 0.65$, $r_{17}^{\Sigma} == 0.26$, $r_{18}^{\Sigma} == 0.84$, $r_{19}^{\Sigma} == 0.69$, $r_{110}^{\Sigma} == 0.55$, $r_{111}^{\Sigma} == 0.73$, $r_{112}^{\Sigma} == 0.73$, $r_{113}^{\Sigma} == 0.67$, $r_{114}^{\Sigma} == 0.98$

To make certain optimum alternative r_{ij*}^{Σ}:

$$\begin{aligned} r_{ij*}^{\Sigma} &= \max\{r_{i1}^{\Sigma}, r_{i2}^{\Sigma}, \ldots, r_{im}^{\Sigma}\} \\ &= \max\{0.79,0.89,0.64,0.74,0.64,0.65,0.26,0.84,0.69,0.55,0.73,0.73,0.67,0.98\} \\ &= 0.98 = r_{114}^{\Sigma} \end{aligned}$$

We thus said that b_{14}, *S. chinensis* (gall), was the optimum alternative with highest antioxidative capacity. We can rank the r_{ij}^{Σ} value to an alternative order in Table 5.

Table 5. The rank of antioxidative capacity in insect galls and host plants leaves based on grey decision making.

Galls and leaves	r_{ij}^{Σ}	Grey decision order
Daphnephila taiwanensis *(gall)*	0.79	4
Daphnephila sueyenae (gall)	0.89	2
Machilus thunbergii (leaf)	0.64	12
Bruggmanniella sp. (gall)	0.74	5
Litsea acuminata (leaf)	0.64	11
Unknown sp. 1 (gall)	0.65	10
Ficus erecta var. beechyana (leaf)	0.26	14
Unknown sp. 2 (gall)	0.84	3
Sycopsis sinensis (leaf)	0.69	8
Pseudoregma bambucicola (gall)	0.55	13
Styrax suberifolia (leaf)	0.73	7
Ceratovacuna nekoashi (gall)	0.73	6
Styrax formosana (leaf)	0.67	9
Schlechtendalia chinensis (gall)	0.98	1

Among these 14 insects galls and host plants leaves, the antioxidative capacity was *S. Chinensis* (gall) > *D. sueyenae* (gall) > unknown sp. 2 (gall) > *D. taiwanensis* (Gall) > *Bruggmanniella* sp. (gall) > *C. nekoashi* (gall) > *S. suberifolia* (leaf) > *S. sinensis* (leaf) > *S. formosana* (leaf) > unknown sp. 1 (gall) > *L. acuminata* (leaf) > *M. thunbergii* (leaf) > *P. bambucicola* (gall) > *F. erecta* var. beechyana (leaf). The insects galls with stronger antioxidative capacity than host plants leaves was observed, except for unknown 1 (gall) and *P. bambucicola* (gall) which showed relatively lower antioxidative capacity (Table 3).

Liu *et al.* (2010) demonstrated that accumulation of ROS at the gall midge attacked site in three isogenic wheat lines [22]. In the galls induced by *Pseudophacopteron* sp. in *Aspidosperma australe*, the ROS were more concentrated in the cells of the inner cortex, next to the nymphal chamber [23]. The nutgall caused by the Chinese sumac aphid, *S. chinensis*, exhibited good antifungal activity and had been used as natural drugs [9]. In our results, *S. chinensis* (gall) performed highest antioxidative capacity, and the insect galls generally exhibited a relatively higher antioxidative function than the leaves (Table 2). It indicated that the process of galls formation can cause oxidative damage of plant cells and therefore, the plants cells, in order to suppress oxidative damage, had high capacity to free radical scavenging.

The first conclusion, polyphenols, flavanoids, anthocyanins, carotenoids and chlorophylls made different contributions to the four antioxidative functions. The contents of polyphenols from galls and their host plants leaves were the most relevant to the antioxidant ability, whereas the content of chlorophylls is the less. After infection, the polyphenols content in galls largely increased, and chlorophyll content is great reduced. The increment of second metabolites when plant tissues attacked by insect, not only used for defending insects but also can be used for helping to reduce oxidizes injury of cells.

The second conclusion, the organism possessed innate defense mechanisms to scavenge free radicals effectively. The galls had enhanced level of antioxidative capacity to balance the increases in free radicals in their bodies, and may be valuable for the use of crude drugs in the future.

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