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Research Article

Study on potential bio-adsorption of Tangerine peel in removal of heavy metals: Pb, Cd and Ni of vegetable coriander

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The dried peel of Citrus reticulata is used as nutraceutical ingredient in dietary supplements and functional and conventional foods.Meanwhile there is a growing demand to find relatively efficient, low-cost and easily ecofriendly and available adsorbents for the adsorbing process of heavy metals, particularly if the adsorbents are the wastes from agricultural or food industries. Recent studies showed that citrus peels have the potential of adsorption of heavy metals. Tangerine Peel as a one of the most common fruit waste materials was chosen for adsorbing the heavy metals from contaminated soil in order to bio-adsorbing Nickel, Cadmium and Lead from soil as well as Vegetable Coriander which is cultivated and finally would be eaten by people. Packed Potting Soil purchased from recognized centers from Tehran market and 40 same vases in the same situation such as light, water, soil, temperature and other conditions were prepared. The seeds of *Coriandrum sativum* (coriander) were being planted directly on the ground after a cold in vases. Meanwhile in 35 vases the Tangerine peel (TP) by 1%, 3%, 5% dry weight percentage were mixed by soil, due to observing the effect of TP as adsorbent and in specified times of 0, 10, 20, 30, 40 days in the leaves of edible vegetable in treated and untreated soils as an edible vegetable has been grown in these soil samples was studied. The soil, leaves and roots of vegetable were digested by wet method according the standard protocol. The data were analyzed using Analysis of Variance, (ANOVA). Results revealed that Tangerine peel has more potential to adsorb Cadmium and lead first days of study ($p<0.001$) and adsorption capacity varied by considering the effects of various parameters like contact time, initial concentrations, pH, and absorbent dose. TP can accumulate high level of Cadmium in a short time and the uptake rate by vegetable edible plant is significantly affected by their concentrations in the contaminated soil ($p<0.05$), but adsorbing Nickel in a few percentage has been done. A contact time of 30 days by 5% Coriander was found to be optimum and after 40 days cadmium contents were not detected. Decreasing lead content after 20 days in treated soil samples by TP was significant.

Keywords: Tangerine Peel, adsorbent, Cadmium, vegetable coriander, soil.

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Introduction

The dried peel of Citrus reticulata is used as nutraceutical ingredient in dietary supplements and functional and conventional foods. The dried peels especially Tangerine peel are used also in traditional herbal medicine. The synergy company uses tangerine peel in its products in relatively small amounts, intended to provide nutrional suppert, compared with the therapeutic or medicinal amounts commonly used in traditional Asian herbal medicine especially in China. The Tangerine peel is dried also as a sweet spicy flavor. The fruits are eaten mostely fresh, preserved whole in syrup, canned or juiced. The essential oil is used for flavoring in the food industry[1-6]. Risk of chronic diseases can be reduced by frequent consumption of fruits and vegetables. A common component of food products is dietary fiber which consists of variety of non-starch polysaccharides such as cellulose, hemi celluloses, pectin, β-glucans, gums and lignin [6]. Studies found in the Journal of Bio Med Central Dermatology show that Tangerine Peel Tea together with black tea can lower the risk of coming down with

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squamous cell skin carcinoma.Tangerine Peel Tea contains salvestrols which promotes cellular death of breast, lung, ovarian and prostate cancer cells while not causing any problems with healthy tissues. Tangerine Peels should be dried with the inner pith attached by just allowing the peel to sit on a counter to dry until hard. Then grind the peel in a coffee grinder into a fine powder which can be added to recipe without adding much flavor. Or a good size piece of dried peel can be added to hot water for making tea which is great tasting either hot or cold. Also the boiled peels can be added to food, shredded into salads, cakes, candy, and used in all kinds of cooking recipes. Tangerine peel can be used in baths to sooth away stress and tangerine oil can be used in vaporizers for stress [7].

Agroindustrial residues, such as those generated in citric juice production plants, have also been used as a substrate for the production of enzymes [8]. Studied the use of orange peel as a substrate and inducer in the production of polygalacturonase by microorganisms and concluded that orange peel is a very good inducer. Mamma and his colleagues in 2008 concluded that it is possible to produce pectinolytic, cellulolytic and xylanolytic enzymes from the fungal strains of the genera Aspergillus, Fusarium, Neurospora and Penicillium and generate multienzyme activity using a simple growth medium consisting of a solid by-product of the citrus processing industry (tangerine peel) [9]. Tangeritin, a bioflavonoid found in tangerine peel, has been shown to strengthen epithelial cells in a manner that inhibits the metastasis of cancer cells [10]. Naturopath Bill Mitchell explained in a lecture that the compound increases the functional integrity of E-cadherin, which is a cell-to-cell adhesive protein found to be deficient in tissue samples of most cancer patients. Based on these results, we can deduce that tangeritin, and its source, tangerine, might be useful as a cancer preventative. The reasoning is simple–about 80 percent of breast cancers start in the epithelial tissue lining the breast ducts, and this bioflavonoid makes the tissue tougher and more resistant. In order to get this benefit you must eat quite a bit of fruit, so the body will have enough left over to store in the tissue [10].

Adsorption is a technique which has been used successfully for the effective removal of heavy metals and chemical compounds from wastewaters generated by the chemical and textile industries. The first step to an efficient adsorption process is the choice of an adsorbent with high selectivity, high capacity and long life. This should also be available in large quantities at low cost [11]. According to Ströher in 2010, these low-cost adsorbents have been investigated in laboratory scale to treat wastewater with varying degrees of efficiency [12]. An example of this class of adsorbents is orange pulp, which has natural adsorption characteristics which resemble those of activated carbon [12]. Solid waste generated during orange juice production can be used to produce ingredients for animal feed (fresh or dried orange pulp). This system includes the most common procedures applied by orange juice production plants regarding the destination of the residues. Most plants make use of almost all the residue generated in the industrial process for the production of animal feed [13]. Moreira et al.in 2004 reported that the solid orange waste obtained could be used as an ingredient for animal feed due to the high content of sugars, fiber and other residual substances [13]. According to data found in the literature, in the production of orange juice the residues correspond to 50% of the raw material. Therefore, since the processing plant investigated in this case processes 16,000 t of oranges day−1, around 8000 t of solid and liquid residues are generated daily. Considering that the liquid residue (“yellow water”) corresponds to 0.5 g 100 g−1(by weight) of the total residue, 40 t of liquid residues and 7960 t of solid residues are generated daily at this plant. Considering that the feed produced from that residue has 10 g 100 g−1 moisture, from 8000 t of solid residues 7200 t of bran can be obtained daily and used as an ingredient for animal feed. Adsorption is considered to be entirely alluring in terms of its performance of removal from dilute waste solutions. Despite the fact that the use of common materials such as activated carbon [14-17], chitosan [18], zeolite, clay [19, 20] is still completely recognized in order to the high adsorption capacity, they lead to overpriced process, too. Along these longs, there is a growing demand to find relatively efficient, low-cost and easily ecofriendly and available adsorbents for the adsorbing process of heavy metals, particularly if the adsorbents are the wastes from agricultural or food industries [21-26]. The new orientation is occurred towards no expensive adsorbents by researches. The dried peel of Citrus reticulata is used as nutraceutical ingredient in dietary supplements and functional and conventional foods.Meanwhile there is a growing demand to find relatively efficient, low-cost and easily ecofriendly and available adsorbents for the adsorbing process of heavy metals, particularly if the adsorbents are the wastes from agricultural or food industries. Recent studies showed that citrus peels have the potential of adsorption of heavy metals. Tangerine Peel as a one of the most common fruit waste materials was chosen for adsorbing the heavy metals from contaminated soil in order to bio-adsorbing Nickel, Cadmium and Lead from soil as well as Vegetable *Coriandrum sativum* (coriander) which is cultivated and finally would be eaten by people.

Material & Methods

Tangerine Peel Sampling Method

30 kilograms Tangerine was purchased from different recognized Tehran market in October 2016. The samples were identified and voucher was deposited in the Herbarium of Faculty of Pharmacy, Pharmaceutical Sciences Branch, Islamic Azad University (IAUPS) Tehran. The fruits were washed extensively under tap water to remove adhering dirt, rinsed with de-ionized water, cut into small spices by small clean cutter and naturally dried in sunlight. Dried peel was grounded using a clean electric mixer, sieved through (Retsch GmbH & CoKG, Germany) mesh size (250 μm) to retain fine particles. To reduce enzymatic browning, the peels were then dipped in a 1% (w/v) citric acid solution for 10 min, drained and dried in an oven at 150°C for 24 hours and homogenized in a blender to utilize in adsorption experiments.

Soil Sampling Method

Packed Potting Soil purchased from recognized centers from Tehran market and 40 same vases in the same situation such as light, water, soil, temperature and other conditions were prepared. The seeds of Coriandrum sativum (coriander) were being planted directly on the ground after a cold in vases. Meanwhile in 35 vases the Tangerine peel (TP) by 1%, 3%, 5% dry weight percentage were mixed by soil, due to observing the effect of TP as adsorbent and in specified times of 0, 10, 20, 30, 40 days in the leaves of edible vegetable in treated and 5 vases as untreated soils were investigated. The leaves of edible vegetable samples which has been grown in these soil samples were studied. The soil leaves and roots of vegetable were digested by wet method according the standard protocol.

Vegetable Sampling Method

Aerial parts of Coriander in every 10 days in companion of Tangerine Peel were separated in 40 days and washed and digested by wet method according the standard protocol for measuring Cadmium, lead and Nickel. Bioaccumulation factors (BAF-s) were calculated for heavy metal content of plant parts (mg/kg) / heavy metal content of soil (mg/kg), for each metal [27].

All Coriander samples were watered each day by tap water (Tehran tap water). The studied samples were managed by the same light situation and some circumstances in order to be compared with each other due to determine the ability of Tangerine Peel in adsorbing Cadmium and Nickel from soil and its potential to avoid transferring heavy metals to coriander and keep safe the eating vegetable. Physical and chemical properties and concentrations of heavy metals in soils, before and after adding Tangerine Peel in the growth period of cultivated Coriander were measured in every 10 days. In order to assess amount of heavy metals in the soil samples, heavy metal concentrations in soils of studied vases were determined by atomic absorption spectrophotometer [28-31].

Heavy metal in Soil, Leaves and Roots of Coriander

The heavy metals: Nickel, Cadmium and lead in the soil, leaves and roots samples such as analyzed before and after treatment in specified times by AAS after digestion of plant materials by AOAC method [32-37]. The plant samples were washed in deionized water dried (24 hrs at 80°C) immediately to stabilize the tissue and stop enzymatic reactions. After drying, samples were ground to pass a 1.0 mm screen using the appropriate Wiley Mill. After grinding, the sample were thoroughly mixed and a 5 to 8 g aliquot withdrawn for analyses and storage [33]. Weighed 0.5 to 1.0 g of dried (80°C) plant material that has been ground (0.5 to 1.0 mm) and thoroughly homogenized and place in a tall-form beaker or digestion tube. Added 5.0 ml concentrated HNO_3 (65%) and cover beaker with watch glass or place a funnel in the mouth of digestion tube and allow to stand overnight or until frothing subsides. Place covered beaker on hot plate or digestion tube into block digester and heat at 125°C for 1 hour. Removed the digestion tube and allowed cooling. Added 1 to 2 ml 30% H_2O_2 and digest at the same temperature. Repeated heating and 30% H_2O_2 additions until digest is clear.

Add additional HNO_3 as needed to maintain a wet digest. After sample digest is clear, removed watch glass and lowered temperature to 80°C. Continued heating until near dryness. Added dilute HNO_3 (10%), and deionized water to dissolve digest residue and bring sample to final volume [31-38].

Evaluation of the essential oils concentrations

The concentrations of the essential oils in the tangerine peels were determined by extraction method using Clevenger apparatus according to international standard protocols [39-40]. In order to extract the essential oils, 50 g of dried powered Tangerine Peel was separately weighed and placed in one liter conical flask and connected to the Clevenger apparatus. 500 ml of deionized water was added to the flask and heated to the boiling point. The steam in combination with the essential oils were distilled into a graduated cylinder for 5 hours and then separated from aqueous layer. The essential oils were diluted by n-hexane at the ratio of to 10 and kept in the refrigerator for further analysis [39-40].

Identification of the essential oils

Due to the fact that the constituents of the essential oils are volatile, therefore their identification requires the application of gas chromatography-mass spectrometry (GCMS). The mass spectra of the samples are compared to the mass spectra of the standards and firm identifications are made. A Hewlett Packard GC model HP-6890 equipped with a 30 meter of 5% phenyl dimethyl siloxan (HP-SMS) capillary column with a Hewlett Packard MS model HP-5973 was employed to carry out the identification of the chemical constituents [40].

Statistical Analysis

The values reported here are means of five values. Data were tested at different significant levels using student t-test to measure the variations between the contaminations in edible vegetable leaves and the dose of bio-adsorbent and contact time parameters before and after treated by tangerine peel. One way analysis of variance (One-ANOVA) was used for data analysis to measure the variations of metal concentrations using SPSS 22.0 software (SPSS Inc, IBM, Chicago, IL).

Table 1- Chemical composition of tangerine peel essential oil

Components	Concentration(%)	Retention Time (min)
Butanal,3-Methyl	2.65	5.51
Oxiranemethanol	4.34	5.84
Propanoic Acid, 2-Methyl-,Methyle	4.99	6.33
Acetic acid	2.04	7.49
n-Methyl-D3-Aziridine	9.07	9.33
4-Hydroxy Cyclopent	1.92	10.11
4-Methyl-1-D1-Aziridine	2.18	11.88
Limonene	28.11	12.87
1,3,5-Triazine-2,4,6-Triamine	2.35	13.95
4-H-Pyran-4-one,2,3-Dihydro-3,5-Dih	2.65	15.66
4-Vinylphenol	2.51	17.24
4-Vinyl-2-Methoxy-Phenol	2.98	19.35
Benzene,1-Chloro-4-Methoxy	1.65	20.97
Cytidine	8.34	21.72
Dodecanoic acid	1.09	23.67
1,4-Diethynylcyclobutene	4.73	24.58
Xanthotoxin	18.40	27.23

Results

The essential oils of citrus tangerine peel was isolated and the concentrations were determined using Clevenger apparatus. The essential oils are complex natural mixtures that can be formed from different chemical compounds with various contents. Constituent compounds of essential oils are in 2 groups with two distinct biosynthetic pathways from each other. Terpenoids derivatives created of the intermediate acetate–malonic acid and aromatic compounds made from shikimic acid and phenylpropanoids. In addition to the above classification, components of essential oils might be classified based on the functional groups present in the structure that include hydrocarbons, alcohols, aldehydes, ketones, esters, phenols, phenolic ethers, oxides and peroxides. The average yields for tangerine peel was 0.91%. Firm identification of the chemical composition of the essential oil of tangerine peel was performed. Seventeen compounds were identified in tangerine peel essential oil (Table 1), where the main components were limonene (28.11%), xanthotoxin (18.40%), n-Methyl-D3-Aziridine (9.07%, cytidine (8.34%), Propanoic Acid, 2-Methyl-,Methyle(4.99%) and 1,4-diethynylcyclobutene (4.73%).

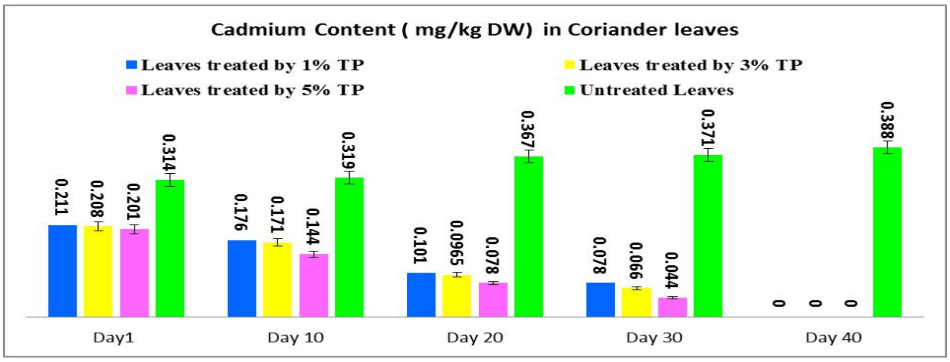

Figure 1. Removal of Cadmium contents from Coriander leaves using tangerine peel (adsorbent dose=1 g/100 g, 3 g/100 g & 5g/100 g), temperature=25 ºC, during 40 days studying.

Results in figures 1 and 2 showed significant differences in Cd up-taking by bio-adsorsbent. The selective potential of certain heavy metals depends on the structure of adsorbent and other factors which affected the percentage of adsorbing such as soil pH and contain exchange capacity and on the distribution of metals among several soil fractions. The fractionation of Pb, Ni and Cd in coriander cultivated control soil and in soils treated by tangerine peel is completely determined due to find out the adsorption ability of heavy metals by agricultural waste in contaminated soil samples. Results showed tangerine peel adsorption for all heavy metals in treated soil were affected significantly by percentage of dried content of adsorbent and the factor of time. Results revealed that Tangerine peel has more potential to adsorb Cadmium during first days of study ($p<0.001$) and adsorption capacity varied by considering the effects of various parameters like contact time, initial concentrations, pH, and absorbent dose. TP can accumulate high level of Cadmium in a short time and the uptake rate by vegetable edible plant is significantly affected by their concentrations in the contaminated soil ($p<0.05$). After 40 days Cadmium content in edible vegetable samples were not detectable.

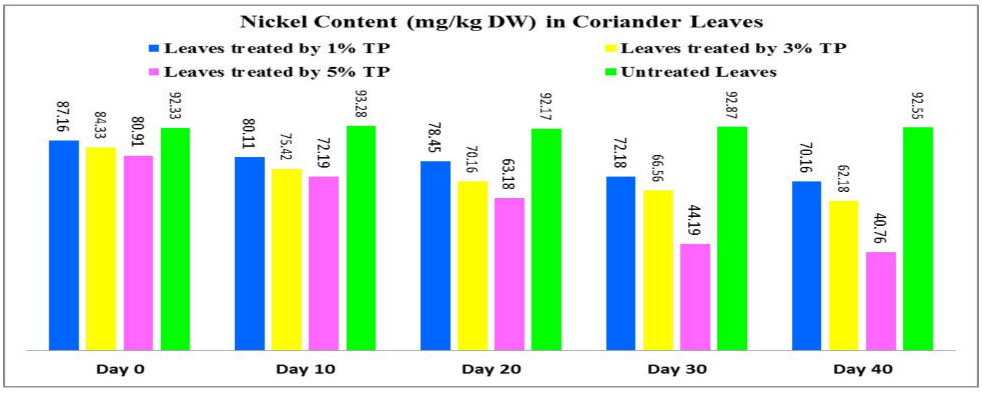

Figure 2. Removal of Nickel contents from Coriander leaves using tangerine peel (adsorbent dose=1 g/100 g, 3 g/100 g & 5g/100 g), temperature=25 ºC, during 40 days studying.

Results from figure 2 revealed that Tangerine peel has less potential to adsorb Nickel during 40 days of study and after 40 days in presence of 5% of TP, reduction of Nickel was significant ($p<0.05$) that approved that adsorption capacity varied by considering the effects of various parameters like contact time, initial concentrations, pH, and absorbent dose. TP can accumulate significant level of Nickel in a long time and the uptake rate by vegetable edible plant is significantly affected by their concentrations of adsorbent in the contaminated soil ($p<0.05$).

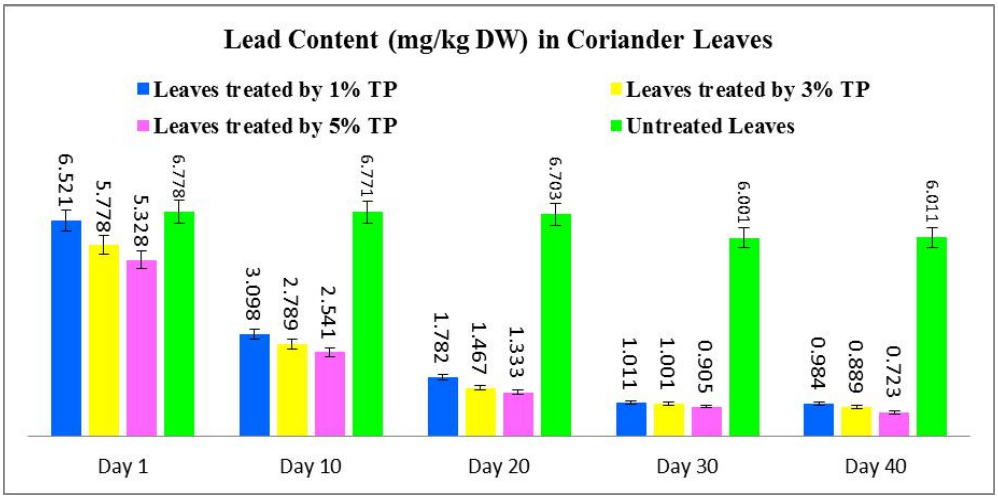

Figure 3. Removal of Lead contents from Coriander leaves using tangerine peel (adsorbent dose=1 g/100 g, 3 g/100 g & 5g/100 g), temperature=25 ºC, during 40 days studying.

Results from figure 3 indicated that Tangerine peel has high potential to adsorb lead during 20 days of study and after 20 days in presence of 5% of TP, reduction of lead was significant ($p<0.003$) that approved that adsorption capacity varied by considering the effects of various parameters like contact time, initial concentrations, pH, and absorbent dose. TP can accumulate significant levels of Lead in a short time and the uptake rate by vegetable edible plant is significantly affected by their concentrations of adsorbent in the contaminated soil ($p<0.001$).

Discussion

The established methods for quantifying the analytical component of the uncertainty is to compare the components that arise from the process of primary sampling, therefore analysis of variance (ANOVA) is applied to estimate the total measurement uncertainty and also to quantify the contributions to that uncertainty which arise from the process of sampling and analysis. ANOVA One-way for statistical comparison between various heavy metals adsorption showed that there is a significant difference in the heavy metals adsorption percentage by the factor of time. The percentage of 5% TP, the best removal rate for Cd, was observed. Similar research studies, but with mango had through FTIR analysis revealed that carboxyl and hydroxyl functional groups were mainly responsible for the adsorption of $Cd2^{+}$. Various metal-binding mechanisms are thought to be involved in this process including ion exchange, surface adsorption, chemisorption and adsorption. Results showed agricultural waste material adsorption for all heavy metals (Cd, Ni and Pb) in treated soil were affected significantly by dried adsorbent content and the adsorbent not only affected on contaminated soil and can adsorb Cd, Ni and Pb after 20-40 days ($p<0.001$) more than other studied but also adding tangerine peel has reduction and rescue effect in taking up heavy metals especially in bio-adsorbing Cadmium and Lead more than Nickel in short time and it keeps edible vegetable safe for eating.

Conclusion

Heavy metals contamination is more prevalent due to inefficient food regulatory policies, inadequate environmental monitoring, and enforcement strategies. The health implications of trace elements and the toxic consequences of heavy metals necessitate effective monitoring of food products to ensure the public health safety. Tangerine peel is an attractive and inexpensive option for the removal of dissolved metals. This study investigated removal yields of Lead, Nickel and Cadmium contents of edible vegetables using tangerine peel. Thus detoxification of heavy metals in plants, a serious issue before being used for food processing and human consumption, finds a better treatment process with TP. Heavy metal related illnesses and chronic degenerative conditions can be avoided in *Coriandrum sativum* (coriander) using tangerine peel treatment in Soil. The responsible organizations should stimulate research to upgrade existing soil treatment by implementing more properly adsorbents especially from food waste industry and agricultural waste materials and demonstrating their reliability to the public.

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Conflicts of Interest

None of the authors have any conflicts of interest associated with this study.

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