

Determining the Extent of Contamination of Vegetables Affected by Tannery Effluent in Ejersa Area of East Shoa, Ethiopia

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Abstract- A field experiment was carried out during 2010/2011 and 2011/2012 years to determine the extent of contamination of vegetables by tannery effluent in Ejersa area of East Shoa, Ethiopia. Five treatments comprised of tannery effluent of different concentrations levels, i.e., 0, 25, 50, 75, 100% applied to six vegetable plants like onion (*Allium cepa* L.), carrot (*Daucus carota* L.), beet root (*Beta vulgaris* L.), Swiss chard (*Beta vulgaris* L.), tomato (*Lycopersicon esculentum* L.) and cabbage (*Brassica oleracea* L.). The treatments were arranged in Randomized Complete Block Design (RCBD) with five effluent concentrations (treatments), each replicated three times. All vegetable plants grown with different effluent concentrations and the effects of different concentrations of effluents were compared to that of normal water (control). Parameters considered to study were heavy metals like (cadmium, chromium, copper, lead, iron, zinc). Recently matured leaf samples were collected during fifty percent maturity and during full maturity. A composite of 20 leaf samples were collected from each plot, then analyzed and compared with those of natural limits and the safe limits of various agencies for vegetables. Swiss chard irrigated with different effluent concentration have been shown to accumulate relatively high concentrations of heavy metals compared to other vegetables included in the experiment. Cd, Cr, Fe and Pb in T₃ (75 % effluent concentration) and T₄ (100% effluent concentration) were recorded higher than the maximum limit for vegetables. Cu and Zn were recorded less than the maximum limits for vegetables. All metal concentrations increased with increasing effluent concentrations. Cabbage irrigated with different effluent concentrations, showed less concentrations of Cu, Fe, Pb and Zn which were found less than the maximum allowable limit for vegetables. Chromium was found to be less than the maximum allowable limit for vegetables. Comparing with other vegetables the concentration of heavy metals was recorded less in cabbage. Continuous monitoring of soil, plant and water quality together with prevention of metals entering to vegetables is prerequisite in order to prevent potential health hazards to human beings.

Index Terms- Contamination, Heavy Metals, Tannery Effluent, Vegetables.

I. INTRODUCTION

Vegetables are important ingredient of human diet that contain essential nutrients and trace elements (Abdullah and Chmielnicka, 1990). Vegetables constitute an important part of human diet since they contain carbohydrates, proteins, as well as vitamins, minerals, trace elements and fibers, and also have beneficial anti oxidative effects (Dastane, 1987). Until recently, however, they did not constitute a major part of the Ethiopian diet, except during the fasting period.

However, since recent years their consumption is increasing gradually, particularly among the urban community. This is due to increased awareness on food value of vegetables, as a result of exposure to other cultures and acquiring proper education (Fisseha Itanna, 2002). Wastewater irrigation results in significant mixing of heavy metal content of agricultural land (Mapanda et al., 2005). The chief cause is the waterways through which heavy metals are leached out of the soil and are taken by the vegetation. If plants decay, these toxic metals are redistributed and as a consequence their enrichment in the agricultural soil occurs. Bioaccumulation, geo accumulation and biomagnifications may result because of entrance of these heavy metals to the ecosystem. Thus long term wastewater irrigation leads to build up of heavy metals in soils and food crops (Khan et al., 2008). Rapid industrialization and urbanization with insufficient environmental monitoring planning often results in discharging of the industrial and sewage waste into rivers and lakes which lead to gradual pollution of our water resources. Many times such wastewater is drained to the agricultural land where this polluted water is used for irrigating crops including vegetables.

Thus polluted effluent water is found to be rich not only in organic matter and nutrients but also in heavy metals like lead, chromium, cadmium, nickel, cobalt etc that finally reach to the soil of agricultural area and leads to food chain contamination as crops and vegetables absorb them from the soil. Heavy metals are not easily biodegradable and it leads to their accumulation in human vital organs causing varying degrees of illness on acute and chronic exposure (Ward et al., 1995). Heavy metal contamination of vegetables cannot be underestimated as these foodstuffs are important components of human diet.

The vegetable farm at Ejersa is among the vegetable farms of the area, where a substantial amount of vegetables are being produced seasonally. The farm is irrigated with the wastewater from the effluent by the tannery and from underground water that may be contaminated by the effluent of the tannery. Before several decades, the water from the river in the area was clean. However, with the increase in the urban population and industrialization, the water has now become contaminated with various pollutants among which are heavy metals. Vegetables grown at contaminated sites could take up and accumulate metals at concentrations that are toxic (Weigert, 1991). In addition they could be contaminated as farmers wash them with wastewater before bringing them to market. The leafy vegetables under this study; namely, cabbage and Swiss chard are usually mixed with potatoes and carrots and cooked to form a special sauce known as "alicha", while tomato is usually cut in pieces, seasoned well and eaten raw as salad (Fisseha Itanna, 2002).

Heavy metal accumulation may pose a direct threat to human health (Turkdogan et al., 2003). Cadmium accumulation may cause bone deformation, kidney damage, anemia, injury of central nervous system and liver disease. Copper toxicity may induce hypertension, coma and sporadic fever. Zinc accumulation causes vomiting and renal damage, whereas hexavalent chromium may induce gastro-intestinal ulceration and cancer (Prabu, 2009). Heavy metal contamination of the food items is one of the most important aspects of food quality assurance (Marshall, 2004; Radwan and Salama, 2006; Wang et al., 2005; Khan et al., 2008). International and national regulations on food quality have lowered the maximum permissible levels of toxic metals in food items due to an increased awareness of the risk these metals pose to food chain contamination (Radwan and Salama, 2006).

II. MATERIALS AND METHODS

The study was conducted during 2010/2011 and 2011/2012 years at the Ethiopia Tannery Share Company (ETSC), one of the largest tanneries of the country found about 90 km East of Addis Ababa in Ejersa area of East Shoa.

The seed of six vegetables onion (*Allium cepa* L., Var. Bombay red), carrot (*Daucus carota* L., Var. nantus), beet root (*Beta Vulgaris* L., Var. Detroit dark red.), Swiss chard (*Beta Vulgaris* L., Var. Fordhook giant), tomato (*Lycopersicon esculentum* L., Var. roma VF) and cabbage (*Brassica oleracea* L., Var. Copenhagen market) vegetable plants were purchased from vegetable seed importers by keeping the higher standard of purity and quality of germination. The effluent is discharged from Ethiopia Tannery Share Company. The effluent was collected in big plastic containers directly from the outlet of the tannery and prepared in different concentrations. The treatments were made by mixing measured amounts of wastewater in T₀ (normal water) i.e. diluted T₁ (25%), T₂ (50%), T₃ (75%) and undiluted T₄ (100%) effluent. The vegetables were grown in three replications on the plot size of (1.2 m x 2 (2.4 m²)) for onion, carrot, beet root, Swiss chard and for tomato & cabbage on 1.8 m x 2 m (3.6 m²). Depending on the space between plants and rows, the total area of the experiment plots was 415.00 m². The Treatments were arranged in Randomized Complete Block Design (RCBD) with three replications.

All cultural practices like:- land cultivation, sowing depth, transplanting, space between plants and rows, hoeing, weeding, fertilizer applications and pesticides applications for all plots were kept uniformly as per recommendations. Vegetable plants were irrigated with normal water against mixture of different effluent concentrations every other days in accordance with the plant requirements through the crop period for full nourishment of the vegetable plants and to keep the soil moist using hand held pouring cans. Recently matured leaf samples were collected from the field irrigated with different effluent concentrations during half maturity (fifty percent flowering) and full maturity. The samples were collected twice during a period of two years to represent the dry and rainy season and divided into leaf and root. A composite of 20 leaf samples were collected from each plot.

The leaf samples were prepared in laboratory, where they were thoroughly washed to remove all adhered soil particles. Samples were cut into small pieces, air dried for 2 days (depending of the plant type) and finally dried at 100 ± 1 °C in an oven for 72 hrs. The sample were ground in warm condition and passed through 1 mm sieve. Samples were analyzed to determine heavy metal accumulation in vegetable plants following the method given by APHA (1998) FAAS (Flame Atomic Absorption Spectrometer) and GFAAS (Graphite Furnace Atomic Absorption Spectrometer) Analyst 600 Perkin Elmer were used to determine heavy metals such as Cd, Cr, Cu, Fe, Pb, and Zn.

The 0.5 gm of well homogenized sample was weighed into a clean silica dish and 0.5 ml of 20% sulphuric acid was added. Thorough mixing of wet samples was done and stirring rod was rinsed with water into clean silica dish. Content of the dish was dried in an oven at 110°C. Then it was heated over a soft flame until all volatile matter was removed. The dish was later transferred to furnace set at 250°C and temperature was raised slowly to 500°C for about 6 to 8 hr. If the ash was not carbon free, 0.5 ml of nitric acid was added and dish was returned to the furnace at 500°C and ashing was done for about 30 min. It was done triplicate. Later 1 ml of nitric acid and 10 ml of water were added to the clean ash and the mixture was heated till the ash was dissolved. The content was qualitatively transferred to a 50 ml volumetric flask. Sample blank solutions were prepared using the same procedures described for the samples. Same quantities of reagents including water were used for sample and blank. All chemicals used were of analytical grade. The 1000 ppm stock solution of each of the metal ion was prepared and required dilutions were made for the purpose of calibration curves. Both sample and blank solutions were analyzed with AAS and concentrations of all metals were determined (MMAF, 2005).

The collected data were subjected to combined statistical analysis of variance (ANOVA) over years using SAS software package 2010. Treatment means that showed significant differences were separated by Duncan's Multiple Range Test (DMRT).

III. RESULTS AND DISCUSSIONS

The data presented in Table 1 shows the effect of tannery effluent with concentration of T₀ (0% normal water, i.e control) and T₁ (25% effluent concentration) was found Cd = 0.001 & 0.12 mg kg⁻¹, Cr = 0.002 & 1.15 mg kg⁻¹, Cu = 0.007, 4.10 mg kg⁻¹, Fe 0.005 & 6.00 mg kg⁻¹, Pb = 0.007 & 0.08 mg kg⁻¹ Zn = 0.009 & 8.80 mg kg⁻¹ which was less than the maximum limit for vegetables. Germination failure was observed at higher effluent concentration of 50,75 and 100% plant was very susceptible to high effluent concentrations, which is T₂ (50% effluent concentration), T₃ (75% effluent concentration) and T₄ (100% concentration), no growth was observed in both two years. This may be attributed to the toxicity caused due to increasing amount of various organic and inorganic compounds present in higher concentration of the effluent. Statistically significant differences between treatments were observed in all metal concentrations.

Table 1. Mean metal concentrations in onion plant irrigated with tannery effluent representing the dry and rainy seasons during 2010/11 and 2011/12 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.001 ^a	0.002 ^a	0.007 ^a	0.005 ^a	0.007 ^a	0.009 ^a
T ₁	0.12 ^b	1.15 ^b	4.10 ^b	6.00 ^b	0.08 ^b	8.80 ^b
CV (%)	3.76	8.76	10.43	11.42	5.55	12.68
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level.

*Source: FAO/WHO-Codex alimentarius commission, 2001

Table 2 shows that, the mean concentration of Cd, Cu, Fe, Pb and Zn in all treatments (T₁ – T₄) were increased with increasing effluent concentrations. The mean concentration of metals in carrot plant however, were recorded less than the maximum limits for vegetables. Chromium was generally the highest concentration in the analyzed samples.

It was also observed the same germination problem in treatment T₁, T₂ and T₃, but in Treatment T₄ (100% effluent concentration) no plant growth was observed in both two year experiments, this may happen due to the susceptibility of the carrot to the highest concentration of tannery effluent. The result showed statistically significant differences between treatments (P<0.05) for all metal concentrations.

Table 2. Metal concentrations in carrot plant irrigated with tannery effluent representing the dry and rainy seasons during 2010/ 2011 and 2011/2012 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.003 ^a	0.002 ^a	0.007 ^a	0.008 ^a	0.005 ^a	0.009 ^a
T ₁	0.07 ^b	1.35 ^b	5.74 ^b	9.50 ^b	0.13 ^b	9.60 ^b
T ₂	0.10 ^c	1.82 ^c	7.50 ^c	11.42 ^c	0.29 ^c	22.46 ^c
T ₃	0.13 ^d	2.42 ^d	15.90 ^d	16.65 ^d	0.64 ^d	38.75 ^d
CV (%)	1.74	3.76	8.73	6.48	3.26	4.32
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level.

*Source: FAO/WHO-Codex alimentarius commission, 2001

The mean metal concentrations in beet root irrigated with different effluent concentrations, shows the mean concentration level of Cd, Cu, Fe and Zn were found less than the maximum allowable or permissible limit for vegetables. Chromium (Cr) and lead (Pb) were found more than the maximum allowable limit for vegetables. All heavy metals concentrations in treatments T₁ – T₃ (25% - 100%

effluent concentrations) were observed in increasing order, which means the concentration of heavy metals increases with increasing effluent concentrations which showed statistically significant differences between treatments (Table 3).

Table 3. Metal concentrations in beet root irrigated with tannery effluent representing the dry and rainy seasons during 2010/11 and 2011/12 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.002 ^a	0.004 ^a	0.005 ^a	0.007 ^a	0.007 ^a	0.005 ^a
T ₁	0.09 ^b	1.23 ^b	0.31 ^b	23.45 ^b	0.12 ^b	6.63 ^b
T ₂	0.12 ^c	1.42 ^c	10.33 ^c	77.32 ^c	0.15 ^c	22.65 ^c
T ₃	0.15 ^d	2.34 ^d	13.27 ^d	194.63 ^d	0.87 ^d	72.14 ^d
T ₄	0.19 ^e	3.32 ^e	31.45 ^e	384.30 ^e	1.22 ^e	90.40 ^e
CV (%)	4.33	5.27	8.68	10.45	4.82	9.64
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level

*Source: FAO/WHO-Codex alimentarius commission, 2001

The mean metal concentration in Swiss chard irrigated with different effluent concentrations have been shown to accumulate relatively high concentrations of heavy metals comparing to other vegetables included in the experiment. In this study the mean concentration of Cd, Cr, Fe and Pb in T₃ (75 % effluent concentration = 0.26, 2.03, 110.51, 0.32 mg kg⁻¹) respectively and T₄ (100% effluent concentration = 0.29, 2.45, 217.10 and 0.43 mg kg⁻¹) respectively, were recorded higher than the maximum limit for vegetables. Cu and Zn were recorded less than the maximum limits for vegetables. All metal concentrations increased with increase of effluent concentrations which showed statistically significant differences between treatments (Table 4).

Table 4. Mean metal concentrations in Swiss chard irrigated with tannery effluent representing the dry and rainy seasons during 2010/11 and 2011/12 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.004 ^a	0.007 ^a	0.007 ^a	0.007 ^a	0.002 ^a	0.002 ^a
T ₁	0.12 ^b	1.34 ^b	8.31 ^b	21.21 ^b	0.14 ^b	12.67 ^b
T ₂	0.20 ^c	1.62 ^c	18.33 ^c	84.32 ^c	0.25 ^c	31.65 ^c
T ₃	0.26 ^d	2.03 ^d	27.41 ^d	110.51 ^d	0.32 ^d	51.13 ^d
T ₄	0.29 ^e	2.45 ^e	33.17 ^e	217.10 ^e	0.43 ^e	69.43 ^e
CV (%)	1.56	3.32	5.67	11.44	1.35	6.42
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level.

*Source: FAO/WHO-Codex alimentarius commission, 2001

Table 5 shows that, the mean concentrations of Cd, Cr, Cu, Fe, Pb and Zn in all treatments (T₁ – T₄) increased with increasing effluent concentrations, even though, the concentration of metals in tomato plant were recorded less than the maximum limit for vegetables. In treatment T₄ chromium (Cr) shows the slightly more than the maximum allowable limit for vegetables. Lead (Pb) was also found more than the maximum limits for vegetables. In Treatment T₄ (100% effluent concentration) no plant growth was observed in both two years, this may happen due to the susceptibility of the tomato plant to the highest concentration of tannery effluent. The result showed statistically significant differences between treatments (P<0.05) for all metal concentrations.

Table 5. Mean metal concentrations in tomato plant irrigated with tannery effluent representing the dry and rainy seasons during 2010/11 and 2011/12 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.006 ^a	0.007 ^a	0.005 ^a	0.005 ^a	0.002 ^a	0.004 ^a
T ₁	0.10 ^b	1.14 ^b	5.31 ^b	34.53 ^b	0.12 ^b	8.63 ^b
T ₂	0.13 ^c	1.92 ^c	12.74 ^c	97.63 ^c	0.28 ^c	32.35 ^c
T ₃	0.18 ^d	2.33 ^d	15.43 ^d	124.51 ^d	0.37 ^d	43.23 ^d
CV (%)	0.94	2.58	4.76	11.66	1.13	5.38
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level.

*Source: FAO/WHO-Codex alimentarius commission, 2001

The mean concentration level of Cd, Cu, Fe, Pb and Zn in cabbage treated with different effluent concentrations were found less than the maximum allowable limit for vegetables. Chromium was found less than the maximum allowable limit for vegetables, which was T₁ = 0.74, T₂ = 1.42, T₃ = 1.83 and T₄ = 2.13 mg kg⁻¹. Comparing with other vegetables the concentration of heavy metals were recorded less. The concentration of heavy metals in all treatments were recorded to increased with increasing effluent concentrations which were significantly difference between treatments (Table 6).

Table 6. Mean metal concentrations in cabbage plant irrigated with tannery effluent representing the dry and rainy seasons during 2010/11 and 2011/12 years

Treatments	Metals content (mg kg ⁻¹) (Mean of three replications at half and full maturity)					
	Cd	Cr	Cu	Fe	Pb	Zn
T ₀	0.005 ^a	0.004 ^a	0.009 ^a	0.005 ^a	0.003 ^a	0.006 ^a
T ₁	0.08 ^b	0.74 ^b	0.31 ^b	23.45 ^b	0.12 ^b	6.63 ^b
T ₂	0.13 ^c	1.42 ^c	10.33 ^c	77.32 ^c	0.15 ^c	22.65 ^c
T ₃	0.15 ^d	1.83 ^d	12.41 ^d	86.51 ^d	0.22 ^d	51.23 ^d
T ₄	0.19 ^e	2.13 ^e	26.30 ^e	103.15 ^e	0.29 ^e	53.64 ^e
CV (%)	0.54	2.46	5.52	8.27	1.22	4.72
Maximum limits for Vegetables*	0.2	2.3	73.3	425.5	0.3	99.4

Means followed by different letters within the same column are significantly different at 5% probability level.

*Source: FAO/WHO-Codex alimentarius commission, 2001

It was observed that all metal contents in all vegetable plants were found to be lowest in treatment T₁ (25% effluent concentration) and highest in T₄ (100% effluent concentrations). Present investigation has clearly shown that the mixture of tannery effluent in higher proportion may cause toxicity during the consumption of the vegetables.

Differences in metal concentrations in vegetables seem to imply that different types of vegetables have different abilities to accumulate metals. In spite of the mechanism involved in the element uptake by root, plants are known to respond to the amount of readily mobile metals in soil. The order of toxic heavy metal concentrations in vegetables vary with toxic metals. Different vegetable species accumulate different metals depending on environmental conditions, metal species and plant available forms of heavy metals (Lokeshwari & Chandrappa, 2006).

Genotypical differences in tolerance and co-tolerance to heavy metals are well known in some species and ecotypes of natural vegetation. Onion, carrot and tomato plants were highly sensitive to the high effluent concentrations, which led to the poor germination and total kill of plants. Cabbage was generally the least accumulator of metals as compared to other vegetables. The mean concentration of Cr was found less than the maximum allowable limit. Cu content in all vegetables was also found less than the maximum limit. Cd accumulation was more in leafy vegetables viz., Swiss chard. The reason for the accumulation is that Cd is

relatively easily taken up by leafy vegetables and also due to the foliar absorption of atmospheric deposits on plant leaves (Midio & Satake, 2003). An earlier study on metal contents of vegetables from Addis Ababa market showed that Swiss chard contained the highest Cd and cabbage the least (Rahlbeck et al., 1999). Leafy vegetables accumulate higher metal contents than others (Al Jassir *et al.*, 2005).

Heavy metals are one of a range of important types of contaminants that can be found on the surface and in the tissues of fresh vegetables. Prolonged human consumption of unsafe concentrations of heavy metals in food stuff may lead to the disruption of numerous biological and biochemical process in the human body (Prabu, 2009). Heavy metal accumulation gives rise to toxic concentrations in the body, while some elements (e.g. arsenic, cadmium, chromium) act as carcinogens and others (e.g. mercury and lead) are associated with development abnormalities in children. The Codex Committee on Food Additives and Contaminants of the Joint FAO/WHO Food Standards Programme, has proposed draft levels for typical daily exposure and theoretical maximum daily intake (TMDI) for some of these metals in vegetables (Codex, FAO/WHO, 2001). Accordingly, the intake of most of the metals constitute less than 10% of the TMDI. Arsenic, chromium, iron and lead are at present of greater concern of health risk than the other metals. Metal concentrations in the vegetables studied would not suffice for determining health implications. This depends also on the dietary pattern of the consumers.

It is because of this, that the intake of metals from the studied vegetables constitutes much less than the TMDI or the provisional tolerable weekly intake (PWTI), which are used to express the exposure of consumers and associated health risk. A recent study on leafy vegetables bought from Addis Ababa market also confirms this (Fisseha Itanna, 1999). However, with increase in vegetable consumption this situation could easily change.

For instance, it has been reported that through the introduction of bio-intensive gardening in some households in Addis Ababa the daily vegetable intake per person has risen from 5gm to 161gm (ENDA, Ethiopia, 2002).

IV. CONCLUSIONS AND RECOMMENDATIONS

From the study it is revealed that, untreated tannery effluent is the main source of pollution to the irrigation with contaminated effluent containing variable amounts of heavy metals leading to increase in concentration of metals in vegetables, which was grown using polluted effluent. Concentration of metals in vegetables will provide baseline data and there is a need for intensive sampling for quantification of results throughout the country. Since cabbage is the least accumulator of metals and metalloids, it may be less risky to eat cabbage at Ejersa than eating Swiss chard from health standpoint. Heavy metals have a toxic impact, but detrimental impacts become apparent only when long term consumption of contaminated vegetables occurs. It is therefore suggested that regular monitoring of heavy metals in vegetables and other food items should be performed in order to prevent excessive build up of these heavy metals in the human food chain.

To avoid entrance of metals into the food chain, tannery effluent should not be discharged into rivers and farmlands without prior treatment. Apart from treating the discharge that enters into the farms, it is also imperative to utilize alternative measures of cleaning up the already contaminated substrates. Continuous monitoring of soil, plant and water quality together with prevention of metals entering vegetables is prerequisite in order to prevent potential health hazards to human beings. Periodical monitoring the rate of contamination and consumption is thus necessary to assess the overall exposure level in the community. Reduced crop contamination and improved safe food can be achieved through, reducing pollution at source, improved vegetable production and post harvest handling and using support for vegetable trading systems to improve food safety.

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