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ANTIOXIDANT INTAKE FROM FRUITS, VEGETABLES, AND BEVERAGES CONSUMED IN LEBANON

by
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AN ABSTRACT OF THE THESIS OF

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Title: Antioxidant Intake from Fruits, Vegetables, and Beverages Consumed in Lebanon

Background: Antioxidants are compounds that are capable of substantially inhibiting or retarding the oxidation of compounds of biological importance including lipids, proteins and DNA in addition to preventing the formation of free radicals (atoms and/or molecules with unpaired electrons). Excessive free radical formation has been linked to premature aging and an increase in the incidence of chronic diseases. Diets rich in antioxidants protect against chronic degenerative diseases and improve longevity. Plant foods including fruits, berries, vegetables, spices, coffee and tea are the major sources of dietary antioxidants. Scant information exists on the dietary intake of antioxidants in developing countries including Lebanon.

Objectives: The study aimed to quantify the phenolic content of fruits, vegetables and beverages frequently consumed in Lebanon for estimating their contribution to the dietary antioxidant intake of the Lebanese population. Another objective of the study is to identify food items whose consumption should be encouraged to promote better quality of life in Lebanon.

Methods: Food consumption data was derived from a National survey conducted in Lebanon between May 2008 and April 2009. The beverages, fruits and vegetables selected for analysis account for at least 85% of the total consumption. Both extractable and non-extractable phenolic antioxidants were assayed for their polyphenol contents by the Folin-Ciocalteu procedure and total antioxidant capacity (TAC) by the trolox equivalent antioxidant capacity (TEAC) and the ferric-reducing ability of plasma (FRAP) assays.

Results: Olives exhibited the highest total phenol (TP) content at 957.9 mg gallic acid equivalents (GAE)/100g fresh weight (FW) followed by instant coffee (375 mg GAE/100 ml), Turkish coffee (369.8 mg GAE/100 ml) and apples (226.8mg GAE/100g FW). The other analyzed foods had lower TP contents ranging in 4.39- 217.8 mg GAE/100g FW. The total dietary phenol intake from the analyzed beverages, fruits and vegetables was 613 mg (GAE)/day with beverages contributing 375 mg GAE followed by fruits at 225 mg GAE and vegetables 13 mg GAE. Further, the largest single source of total phenols (TP) consumed (per day) in Lebanon was black tea (152 mg GAE),

followed by Turkish coffee (120mg GAE), instant coffee (99.4 mg GAE), apples (72 mg GAE), olives (64 mg GAE) and grapes (50.3 mg GAE).
The TAC determined by FRAP and TEAC ranged between 0.17-120.6 mmol Fe^{2+}/L or kg and 0.157-45.1 mmol Trolox/ L or kg, respectively. Instant coffee and olives showed the highest AC in the tested items while cucumbers were the lowest. Both antioxidant tests correlated highly with the TP content and with each other ($P < 0.01$), thereby suggesting that the phenolic compounds are responsible for the antioxidant activity of the tested commodities.
Instant coffee, black tea, apples, Turkish coffee, grapes and olives are the main contributors to the total dietary AC in Lebanon with no significant differences being observed in dietary AC between the TEAC and FRAP assay results (paired T test, $p > 0.05$).
The Content of non-extractable proanthocyanidins ranged between 0.59 and 62.9 mg PA/100g FW in the analyzed fruits and vegetables. Olives had the highest NEPA content followed by apples and watermelon.

Conclusion: Findings from this study highlight the importance of increasing the Lebanese vegetable and fruit daily intakes to the value recommended by FAO/WHO 400 g/ day to increase phenol intakes in Lebanon. Moreover, higher olive and apple intakes, especially black olives and red-skinned apples in view of their high AC, are recommended as a further measure for increasing antioxidant intakes. Also daily intake of coffee (Turkish and instant), below the caffeine daily limit, should be encouraged, Findings from this study also highlight the need for introducing cucumber varieties that express higher TP than those cultivated in the country. Further studies that investigate the relationship between antioxidant intake and factors including: age, gender, socioeconomic status and geographic region are needed to inform better-customized recommendations.

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ILLUSTRATIONS

TABLES

ABREVIATIONS

µg	Microgram
mmol	millimole
AC	Antioxidant Capacity
DF	Dietary Fiber
DHQ	Diet History Questionnaire
DW	Dry Weight
EPP	Extractable Polyphenols
FBS	Food Balance Sheet
FC	Folin-Ciocalteu
Fe^{2+}	Ferrous
FFQ	Food Frequency Questionnaire
FRAP	Ferric Reducing Antioxidant Power
FW	Fresh Weight
GAE	Gallic Acid equivalent
HAT	Hydrogen Atom Transfer
HCL	Hydrochloric Acid
HLB	Hydrophilic Lipophilic Balance
HPP	Hydrolysable Polyphenols
MACAN	Macromolecular Antioxidants
NEPA	Non Extractable Proanthocyanidins
NEPP	Non Extractable Polyphenols
PA	Proanthocyanidins
PP	Polyphenols

RNS	Reactive Nitrogen Species
ROS	Reactive Oxygen Species
SET	Single Electron Transfer
TAC	Total Antioxidant Capacity
TEAC	Trolox Equivalent Antioxidant Capacity
TPC	Total Phenol Content

CHAPTER I

INTRODUCTION

It has long been observed that diet has an important role in either enhancing or preventing diseases (Cantin *et al.,* 2009). Recent research strongly supports the presence of a link between diets that are rich in antioxidant compounds and prevention of various chronic diseases (Pe´rez-Jime´nez *et al*., 2013). In the last few years, antioxidants have been recognized as an indispensible nutrient protecting our bodies from oxidative cell damage that is associated with several conditions including Alzheimer's disease, cancer, heart disease and chronic Inflammation. Recent findings highlight the vital role of antioxidants in health especially that free radical reactions have been involved in many human pathological conditions including neurodegenerative disorders, cardiovascular diseases, pulmonary disorders, some autoimmune diseases, several renal disorders and gastrointestinal diseases (Rajendra *et al.*, 2014).

Free radicals are atoms or molecules that have unpaired electrons making them unstable and highly reactive. In most cases, free radicals initiate a change in other molecules by abstracting a hydrogen atom from C–H, N–H, or O–H bonds within biomolecules or an electron from a metal or metal complex to attain stability. Consequently, the molecule that loses its electron would become a free radical itself capable of starting a chain reaction cascade leading to cell damage (Zampelas & Micha, 2015; Phaniendra & Periyasamy, 2015). Although non-radical species, also called oxidants, such as hydrogen peroxide (H_2O_2), ozone (O_3), singlet oxygen ($^1O_2$), hypochlorous acid (HOCl), are more stable than free radicals and less reactive, they can easily lead to free radical reactions (Pham-Huy *et al.,* 2008).

Reactive oxygen species (ROS) such as hydroxyl (OH•), superoxide ($O_2•^-$) and peroxyl (ROO•), and reactive nitrogen species (RNS) including nitric oxide (NO•) and nitrogen dioxide (NO_2•), are common compounds in cellular metabolism and also produced in high levels in organisms exposed to environmental stresses (Pham-Huy *et al.,* 2008). These species are generated in the cell (through enzymatic or non-enzymatic reactions), produced endogenously from immune cell activation, inflammation, mental stress, excessive exercise, ischemia, infection, cancer, aging or exogenously from air

and water pollution, cigarette smoke, alcohol, heavy or transition metals (Cd, Hg, Pb, Fe, As), certain drugs industrial solvents, cooking (smoked meat, used oil, fat) and radiation. After entering the body, these exogenous compounds are decomposed or metabolized into free radicals (Pham-Huy *et al.,* 2008).

At low and moderate concentrations, ROS and RNS are involved in many physiological processes as immune function (defense against pathogenic microorganisms), maintaining cell homeostasis and mitogenic response (Phaniendra & Periyasamy, 2015) and in the process of phagocytosis where phagocytes release free radicals to destroy invading pathogenic microbes as part of the body's defense mechanism against disease (Pham-Huy *et al.*, 2008).

However, excess production of ROS/RNS along with a deficiency of enzymatic and non-enzymatic antioxidants causes oxidative or nitrosative stress with damaging effects on certain biomolecules (Phaniendra & Periyasamy, 2015). Among these biomolecules, lipids, proteins and DNA/RNA are the main targets of oxidative stress and the resulting changes in these molecules lead to an increase in the probability of mutagenesis (Hussain et al., 2016). Radicals can damage cell membranes and lipoproteins through lipid peroxidation resulting in the formation of cytotoxic and mutagenic compounds including malondialdehyde and conjugated dienes. Moreover, proteins can be affected by three ways: free radical mediated cleavage (e.g. enzyme proteolysis), changes in specific amino acids and formation of cross-links through reaction with lipid peroxidation products. These changes affect enzymes activity, receptors, membrane transport and signal transduction that lead to aging (Lobo *et al.,* 2010). As for DNA, attacks by free radicals can cause damage and mutations; the DNA can be repaired through certain enzymes and/or antioxidants (Pham-Huy *et al.,* 2008). Furthermore, ROS/RNS overproduction in tissues for long periods of time, if not treated, can cause irreversible damage to cells inducing somatic mutations, cell death by necrotic and apoptotic processes, precipitates chronic degenerative diseases as well as some acute pathologies (Hussain *et al.,* 2016; Pham-Huy *et al.,* 2008).

To counteract oxidative stress and prevent its harmful effects, suitable levels of antioxidants must be maintained. Antioxidants are stable molecules that can neutralize excess free radicals and reduce their capacity to damage tissues thereby preventing

diseases. Antioxidants can be produced endogenously by the body or supplied exogenously by diet (Pham-Huy *et al.*, 2008).

Endogenous antioxidants can be divided into enzymatic (e.g. superoxide dismutase, catalase, glutathione peroxidase and glutathione reductase) and non-enzymatic antioxidants. The non-enzymatic antioxidants can be classified as metabolic antioxidants, produced through metabolism, and nutrient antioxidants that must be supplied exogenously through foods or supplements. Nutrient antioxidants include vitamins E and C, carotenoids, trace metals (selenium, manganese, zinc), and polyphenols (Pham-Huy *et al.*, 2008).

Phenols and polyphenols are widely distributed secondary metabolites in the plant kingdom. They are characterized by the presence of one or more phenolic rings in their chemical structure and are classified to different subgroups according to their stability, bioavailability, and physiological functions in relation to human health. More than 8000 phenolics have been reported in fruits, vegetables, grains and herbs (Zampelas & Micha, 2015).

Antioxidants act in several ways including scavenging of radicals, donating hydrogen and electron, decomposing peroxides, inhibiting enzymes, quenching singlet oxygen and chelating metals. Both enzymatic and non-enzymatic antioxidants exist in the intracellular and extracellular environments and play important roles in the detoxification of ROS (Lobo *et al.*, 2010). Antioxidants get oxidized upon neutralizing the free radical thereby necessitating the restoration of antioxidant resources in the body (Pham-Huy *et al.*, 2008). Accordingly, antioxidant intakes (exogenous source) play important roles in helping endogenous antioxidants in mitigating oxidative stress. Nutrient antioxidant deficiency is one of the causes of numerous chronic and degenerative pathologies and diets rich in antioxidants have many benefits to human health including protection against chronic degenerative diseases and improvements in longevity (Pham-Huy et al., 2008).

Plant foods including fruits, berries, vegetables, spices, coffee and tea are the major sources of dietary antioxidants. Scant information exists on the dietary intake of antioxidants in developing countries including Lebanon. Therefore, the purpose of this

study was to evaluate the relative contribution of the most commonly consumed fruits, vegetables and beverages in Lebanon to the antioxidant capacity of the population's diet and the contribution of these commodities to the dietary intake of antioxidants by the Lebanese population. The study also provides information on the major sources of dietary antioxidants in Lebanon and potentially helps consumers in increasing their antioxidant intake thereby contributing to the promotion of better quality of life in Lebanon.

CHAPTER II

LITERATURE REVIEW

It has long been observed that diet has an important role in either enhancing or preventing diseases. In recent times, diet and human well-being have received increasing attention (Cantin *et al*., 2009). Recent research strongly supports the presence of a link between diets that are rich in antioxidant compounds and prevention of various chronic diseases. Many studies showed an inverse relationship between total dietary antioxidant intake and inflammation, ischemic stroke, alterations in endothelial function and gastric Cancer (Pe´rez-Jime´nez *et al*., 2013). Furthermore, adherence to a Mediterranean diet, which increases total dietary antioxidant intake and plasma total antioxidant capacity (TAC) (Kolomvotsou *et al., 2013*), results in reduced incidence of cardiovascular, cancer, Parkinson's, and Alzheimer's disease (Dilis and Trichopoulou, 2010). Moreover, there is increasing scientific evidence on the existence of a direct relationship between damage caused by reactive oxygen species (ROS) and reactive nitrogen species (RNS) on the molecular and cellular levels and the incidence of chronic degenerative diseases including cancer and cardiovascular disease (Saxena *et al.*, 2007).

A. Free Radicals

Reactive oxygen and nitrogen species (ROS, RNS) are small molecules that have single unpaired electron which makes them highly reactive. ROS are grouped in to two categories:

1) Free oxygen radicals including superoxide ($O_2^{\bullet -}$), hydroxyl radical ($^{\bullet}OH$), nitric oxide ($NO^{\bullet}$), organic radicals ($R^{\bullet}$), peroxyl radicals ($ROO^{\bullet}$), alkoxyl radicals ($RO^{\bullet}$), thiyl radicals ($RS^{\bullet}$), sulphonyl radicals ($ROS^{\bullet}$), thiyl peroxyl radicals ($RSOO^{\bullet}$) and disulphides (RSSR); 2) Non radical ROS such as hydrogen peroxide (H_2O_2), singlet oxygen ($^1O_2$), ozone/trioxide (O_3), organic hydroperoxides (ROOH), hypochloride (HOCl), peroxynitrite (ONO^-), nitrosoperoxycarbonate anion ($O=NOOCO_2^-$),

nitrocarbonate anion ($O_2NOCO_2^-$), dinitrogen dioxide (N_2O_2), nitronium (NO_2^+) and highly reactive lipid- or carbohydrate-derived carbonyl compounds (Liou & Storz, 2010). These molecules are produced continuously in the body from normal cellular metabolism and are active in immune response or signaling pathways and they can be controlled within normal limits (Birben *et al*., 2012).

The major targets of ROS and RNS are the unsaturated fatty acids in lipid membranes causing peroxidation that consequently results in loss of membrane fluidity, receptor alignment and lysis of the cells. Moreover, certain aldehydes that arise from degradation of polyunsaturated fatty acids can cause cross linking in lipids, proteins, and nucleic acids. The free radical damage does not only alter lipids but also affects proteins and sulfur containing enzyme through cross linking and denaturation. In addition, mutations in the DNA, that may be carcinogenic, can occur from free radical attach on nucleic acids, and alterations in cellular receptor functions comprising those associated with neurotransmitter and hormonal responses can be precipitated through carbohydrate damage (Landete *et al*., 2013).

High concentrations of ROS and RNS have negative effects on cell components leading to a condition known as "oxidative stress". Put differently, oxidative stress results from the imbalance between oxidants and antioxidants (Qureshi *et al., 2014*). Modern lifestyle patterns and other factors including psychological stress, alcohol consumption, medication, pollution and obesity could lead to the development of abnormally high levels of oxidative stress (Makhlouf *et al.,* 2011). Many studies showed that oxidative stress causes many pathological conditions including diabetes, neurological disorders, atherosclerosis, hypertension, ischemia, idiopathic pulmonary fibrosis, cancer, asthma, acute respiratory distress syndrome and chronic obstructive pulmonary disease. Additionally, it has been claimed that the accumulation of free radical damage over a period of time leads to the aging of the cell as proposed by the free radical theory of aging (Birben, E *et al.,* 2012).

As discussed, because of the insufficiency of the endogenous defense mechanisms present in the body to prevent completely the oxidative damage, exogenous sources are needed such as dietary antioxidants.

B. Antioxidants and Polyphenols

Antioxidants are substances that naturally occur in plants. In addition to conferring pigmentation and certain taste attributes (e.g. astringency) to fruits and vegetables, antioxidants have many health promoting benefits (Celia *et al.,* 2009). At the molecular level, antioxidants are molecules that are capable of substantially inhibiting or retarding the oxidation of other compounds by removing free radical intermediates through getting oxidized themselves. Dietary antioxidants include vitamins A, C, and E and polyphenols. Even though many minerals and trace elements (e.g. Cu, Fe, Mn, Se and Zn) are essential for antioxidant enzyme activity, they are not considered antioxidants (Landete *et al.,* 2013).

Major sources of dietary antioxidants are fruits and vegetables which contribute to nearly 90% of antioxidant activity in the human diet (Landete et al., 2013).The World Health Organization (WHO) recommends eating five servings of vegetables and fruits daily for a healthy diet (Lee *et al.,* 2017). Among the antioxidants, polyphenols (PP) are the mostly consumed contributing to 90% of dietary antioxidant intake and exhibiting higher *in vitro* antioxidant capacity than vitamins and carotenoids (Pérez-Jiménez *et al.,* 2013). The dietary intake of PP is much higher than all other antioxidants being 10× higher than that of vitamin C and 100× times than that of both vitamins E and A (Landete *et al.,* 2013). Due to structural diversity, lack of standardized analytical methods, variation of content in particular food stuff and other reasons, it is extremely hard to estimate the average daily intake of PP. Most authors refer to the data published 25 year ago where a daily consumption of at least 1-2 g of polyphenols including flavonoids has been recommended but the methods used to obtain this result were not detailed (Scalbert & Williamson, 2000; Cieślik *et al.,* 2006). In view of plants being rich sources of polyphenols, beverages that are mainly composed of plant extracts, such as tea and coffee, are also good sources of antioxidants (Fukushima *et al.,* 2009).

Chemically, PP are made up of more than one phenol moiety per molecule (Landete *et al.,* 2013).They can occur either in a non-conjugated form (aglycone) or conjugated with amines, sugars, lipids, carboxylic acids or even other phenols (Pinto and Santos, 2017).

Polyphenols are found in both the edible and non-edible parts of the plant where they retard oxidative degradation of lipids thereby enhancing the quality and nutritional

value of the food (Kähkönen *et al.,* 1999). The antioxidant activity of PP is mostly modulated by their redox properties which allow them to act as reducing agents, hydrogen donators and singlet oxygen quenchers in addition to their metal chelation potential (Kähkönen *et al.,* 1999).

Polyphenols are grouped into flavonoids, phenolic acids and tannins with subclasses within each group (Landete *et al.,* 2013; Ovaskainen *et al.,* 2008).

1. Flavonoids

Chemically, flavonoids are composed of two aromatic rings linked through a heterocyclic pyrone ring (Landete *et al.,* 2013; Ovaskainen *et al.,* 2008). More than 5,000 individual flavonoids have been isolated and identified (Liu, 2013).
The flavonoids comprise flavonols, flavones, flavanones, isoflavonoids, catechins and anthocyanidins which differ in the configuration of the connecting pyrone ring. The most common flavonoids in the diet are flavonols (quercetin, kaempferol, and myricetin), flavones (luteolin and apigenin), flavanols (catechin, epicatechin, epigallocatechin, epicatechin gallate, and epigallocatechin gallate), flavonones (naringenin), anthocyanidins (cyanidin and malvidin), and isoflavonoids (genistein and daidzein) (Liu, 2013). It has been reported that flavonoids can hinder the action of a wide range of enzymes including lipoxygenase, cyclooxygenase, monooxygenase, xanthine oxidase, mitochondrial succinoxidase and NADH-oxidase, phospholipase A2, and protein kinases. These inhibitory effects are believed to originate from their antioxidant properties in addition to their ability to protect against iron induced free radical reactions including those generated at the enzyme's active site (Guohua *et al.,* 1997). The human intake of all flavonoids has been estimated to range between 100 to 650 mg/day whereas that of the two major subclasses, flavonols and flavones, to be 23mg/ day (Liu, 2013).

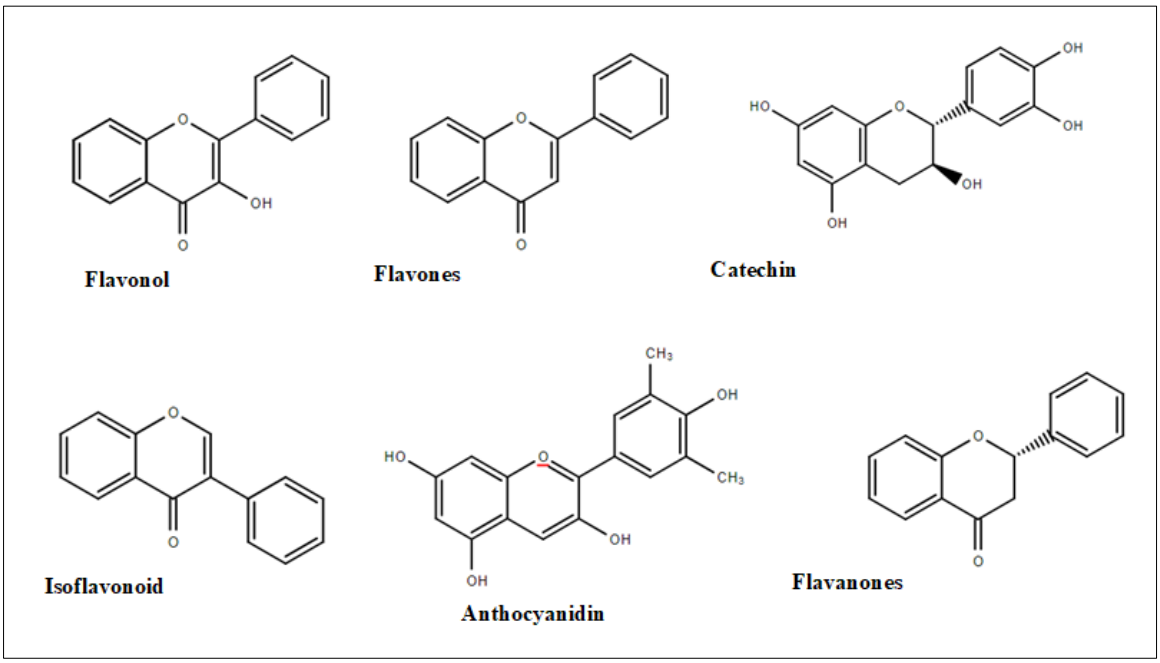

Figure 1. Structures of flavonoids subclasses

2. Phenolic acids

Phenolic acids occur in the form of esters, glycosides or amides and rarely exist in free form. The subclasses of phenolic acids differ in the number and location of the hydroxyl groups present on the aromatic ring (Khoddami *et al.*, 2013). Phenolic acids can be divided to hydroxybenzoic acids and hydroxycinnamic acids. Hydroxybenzoic acids are usually attached to different molecules and are part of complex structure such as lignins, hydrozable tannins, cell walls and proteins. They contain *p*-hydroxybenzoic, gallic acids, syringinc, protocatechuic, and vanillic acids. While hydroxycinnamic acids include *p*-coumaric, ferulic, caffeic, and sinapic acids and are also present in bound form being attached to cellulose, lignin, proteins and cell wall via ester links. (Liu, 2013). The main groups of cell wall phenolics are lignin and hydroxycinnamic acids, being vital for plant growth due to their protective effects against stresses such as wounding, UV radiation and infections (Khoddami *et al.*, 2013).

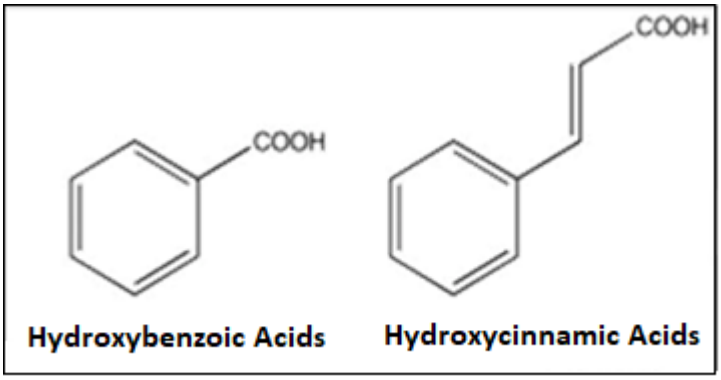

Figure 2. Basic structure of Phenolic acids (Khoddami *et al.*, 2013)

3. Tannins

Tannins are high molecular weight PP comprising the condensed tannins (such as proanthocyanidins PA) and hydrolysable tannins consisting of gallotannins and ellagitannins (Ovaskainen *et al.*, 2008). When ingested tannins can affect utilization of proteins and iron by forming insoluble complexes with both of them and contribute positively to the antioxidant status through participation in redox reactions (Hartzfeld *et al.*, 2002).

Condensed tannins or proanthocyanidins (PA) are a mixture of oligomeric and polymeric dietary polyphenols made up of flavan-3-ol and flavan-3,4-diols that are found extensively in plant foods. In addition to imparting characteristic sensory attributes to foods such as astringency, bitterness, aroma and color, these compounds offer protective effects against gastrointestinal disorders and the progression of chronic diseases (Pérez-Jiménez *et al.*, 2009).

The hydrolysable tannins are made up of a polyol core, often glucose, esterified to gallic acid and exhibit extensive variations in their degrees of esterification and cross linking (Hartzfeld *et al.*, 2002).

C. Determination of food polyphenols

Polyphenols are divided, on the basis of their solubility, into extractable polyphenols (EPP) and non-extractable polyphenols (NEPP). The EPP fraction of food comprises the phenols which dissolve in aqueous solutions of methanol, ethanol or acetone while the NEPP fraction refers to the phenols present in the residue that remains after extraction of EPP. Although the NEPP part in foods may be even higher than that of the EPP

fraction, and although both contribute to the presumed health benefits, it was frequently ignored in research work on antioxidant capacity of foods (Pérez-Jiménez *et al.,* 2013). The extractable polyphenols comprise low molecular weight compounds such as extractable proanthocyanidins (PA) and hydrolysable tannins along with flavonoids, phenolic acids, stilbenes, lignans and others (including tyrosols and alkylresorcinols). In contrast, the non-extractable polyphenols include macromolecules (high molecular weight proanthocyanidins or non-extractable proanthocyanidins NEPA), or low molecular weight compounds (hydrosable polyphenols HPP) such as phenolic acids which are associated with macromolecules mainly the polysaccharide constituents of dietary fiber (DF) and protein (Pérez-Jiménez *et al.,* 2015; Pérez-Jiménez *et al.,* 2013). These non- extractable fractions interact with polysaccharides and proteins through hydrogen bonding, with hydrolysable tannins via hydrophobic interactions and with phenolic acids by covalent bonding. Additionally, a fragment of EPP can be attached with the NEPP fraction and associated with dietary fiber and may be considered one of its elements (Pérez-Jiménez *et al.,* 2013).

The NEPP are also called macromolecular antioxidants (MACAN) due to being either polymeric polyphenols or single polyphenols linked to macromolecular food components that are accessible and bioavailable only in the large intestine (Pérez-Jiménez *et al.,* 2015). The ingested NEPP fractions reach the colon unaffected by mastication, the highly acidic environment of the stomach or digestive enzymes in the small intestine. After reaching the colon, the colonic micro-flora carries out fermentation of these fractions with subsequent release of several bioactive and absorbable metabolites or non- absorbable metabolites that remain in the colonic lumen (Pérez-Jiménez *et al.,* 2009; Pérez-Jiménez *et al.,* 2013). These metabolites are found to neutralize the effect of dietary pro-oxidants and produce an antioxidant environment in the colon offering many health benefits including a chemo-preventive effect for colorectal cancer (Arranz *et al.,* 2009).

The phenols content of EPP are usually quantified with the Folin-Ciocalteu (FC) procedure. The FC reagent contains phosphor-tungstic acid ($H_3PW_{12}O_{40}$) and phosphomolybdic acid ($H_3PMo_{12}O_{40}$) as oxidants which are reduced by the oxidized phenolic hydroxyl groups to produce tungsten oxide (W_8O_{23}) and molybdenum oxide (Mo_8O_{23}) that are blue in color. This reaction takes place under alkaline condition in the

presence of sodium carbonate. The blue color reflecting polyphenols is measured spectrophotometrically at 765 nm and is usually expressed as gallic acid equivalent (GAE) or catechin equivalent (Georgé *et al.*, 2005). However, the FC reagent can react with non-phenolic compounds that have reducing power leading to an overestimation of the polyphenol content of assayed samples. The interfering substances in foods most commonly include ascorbic acid, some sugars and amino acids. The interfering substances are usually removed from the phenol-containing extracts by solid phase extraction on Oasis HLB (hydrophilic-lipophilic balance) cartridges which eliminates the water soluble reducing substances from the extracts. The total polyphenol content is calculated by subtracting the quantities of the interfering substances from the apparent total phenol contents of the samples' extracts (Brat *et al.*, 2006; Georgé *et al.*, 2005). The NEPP fraction present in the residue after extraction of the EPP is divided, for analytical purposes, to non-extractable proanthocyanidins (NEPA) and hydrolysable polyphenols (HPP).

For quantification of the NEPA fraction, the residue is subjected to hydrolysis either chemically, which is most frequently used, or enzymatically in order to break down the polymer.

Chemical hydrolysis entails treating the residue with butanol, hydrochloric acid (HCl) and iron (Fe) solution to break the interflavan linkage in the proanthocyanidins. This converts the polymeric proanthocyanidins to red anthocyanins which are measured spectrophotometrically at 555 nm (Pérez-Jiménez *et al.*, 2009).

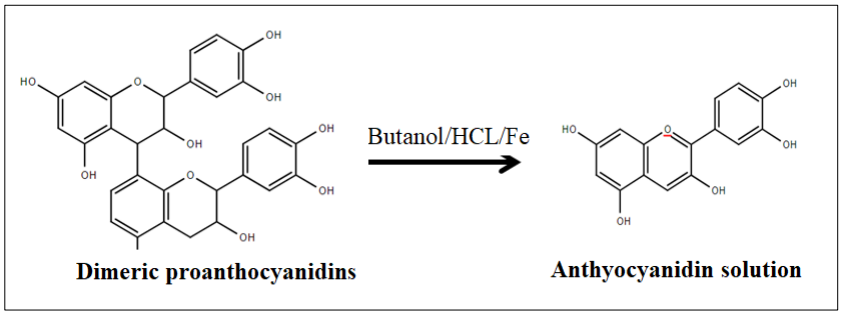

Figure 3. Hydrolysis of dimeric proanthocyanidins in an acidic medium (Pérez-Jiménez *et al.*, 2009)

Moreover, for quantification of HPP the residue of EPP is hydrolyzed using methanol and sulphuric acid to releases methyl gallate from the hydrolysable tannins

(gallotannins and ellagitannins) and then oxidized by KIO_3 at pH 5.5 to form a red colored product that is determined spectrophotometrically. A slower reaction would take place oxidizing this red compound to yellow product. This yellow product is usually yielded when the tested sample contains phenolics other than hydrolysable tannins (Hartzfeld *et al.,* 2002).

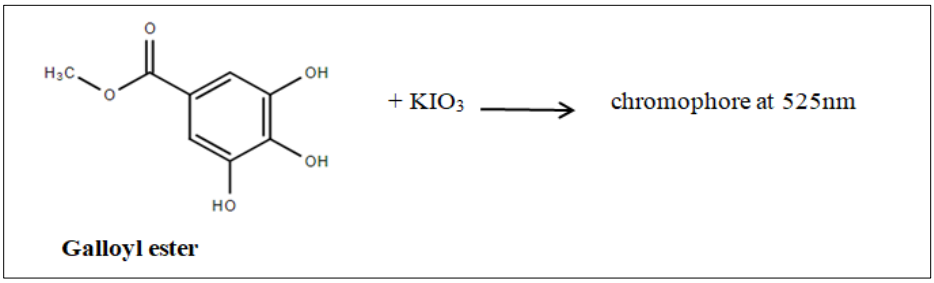

Figure 4. Hydrolysis of hydrolysable tannins in an acidic medium (Hartzfeld *et al.,* 2002)

D. Total Antioxidant Capacity

Since there are many antioxidant compounds in plant foods, it is impractical to measure each separately. For this reason many assays were developed to measure the total antioxidant capacity of foods and biological samples (Guo *et al.,* 2003). The total antioxidant capacity (TAC) measures the cumulative ability of all antioxidants present naturally in a sample of food to scavenge free radicals. The TAC in plant foods is affected by a number of factors including degree of ripening, conditions of storage after harvest such as time, temperature, atmosphere.. , processing procedure including temperature, time and other variables. The most commonly used assays to measure the antioxidant capacity of foods are the ferric reducing antioxidant power (FRAP), ABTS (2,2'-azino-bis(3-ethylbenzthiazoline-6-sulphonic acid), or Trolox equivalent antioxidant capacity (TEAC), the 2,2-diphenyl1-picrylhydrazyl (DPPH) and the oxygen radical absorbance capacity (ORAC). These assays depend on the two major routes through which antioxidant molecules exert their action specifically hydrogen transfer (HAT—hydrogen atom transfer) and electron transfer (SET—single electron transfer) (Pérez-Jiménez *et al.,* 2008).

These two mechanisms quench radicals either by transferring hydrogen atom and recombining with the radical or giving electron to reduce and stabilize these species. These mechanisms are shown below (Schaich *et al.*, 2015).

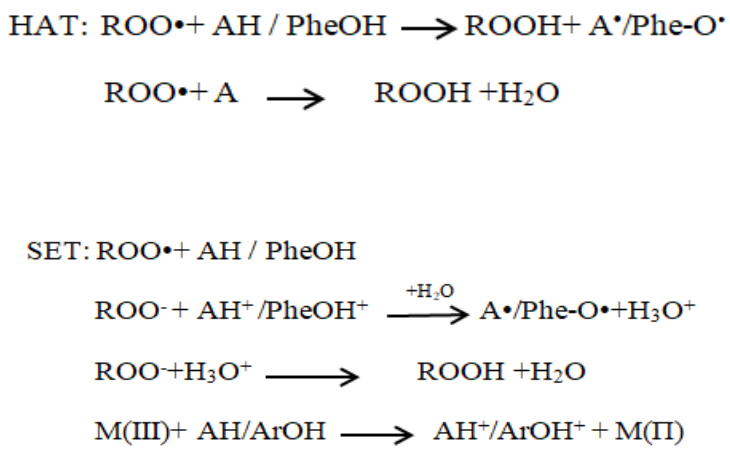

Figure 5. Antioxidants main mechanisms for Quenching free radicals where AH = any antioxidant with donatable H, PheOH = phenol or polyphenol, M = redox-active metal. (Schaich *et al.*, 2015)

Although both assays have the same goal, they differ in kinetics and the solvent used, pH and other conditions. SET is rapid, independent of diffusion, and the rate increases with pH because ionization increases the electron's availability. In contrast, HAT is slowed by diffusion, doesn't rely on pH, strongly enhanced by water and inhibited by hydrogen bonding solvents such as alcohols (Schaich *et al.*, 2015).

Since different antioxidants exert their actions through different mechanisms, no single method can assess the TAC of foods and, accordingly, more than one assay should be used to evaluate the diverse mechanisms of antioxidants' actions. TAC assays based on HAT include the oxygen radical absorbance capacity assay (ORAC) assay and the most commonly used methods utilizing SET are the ferric-reducing Antioxidant power (FRAP) and 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox) equivalent antioxidant capacity (TEAC/ABTS) assay. Moreover, the 2,2'-azino-bis-3-ethylbenzthiazoline-6-sulphonic acid (ABTS) assay and the 1,1-diphenyl- 2-picrylhydrazyl (DPPH) assay that are usually classified as SET methods are also

deactivated either by radical quenching via HAT or by direct reduction through SET mechanisms (Apak *et al*., 2016, 2017; Pérez-Jiménez *et al*., 2008).

ABTS, DPPH and ORAC measure the free radical scavenging capacity; however, the FRAP assay measures the antioxidants' ability in the tested sample to reduce metals. Additionally, ORAC requires a fluorimeter which is not commonly available in many laboratories while the three other assays are performed with a UV spectrophotometer. Concerning the time needed for each it is about 7 min in ABTS, 30 min for FRAP and 30-40 min for ORAC. Moreover, FRAP, ORAC and ABTS usually measure AC of hydrophilic compounds. Some modifications have been applied to the ORAC and ABTS assays in order to determine AC of lipophilic compounds (Pérez-Jiménez *et al.*, 2008).

١.*Ferric-Reducing Antioxidant Power (FRAP)*

FRAP is a simple assay, gives fast and reproducible results which are linearly related to the molar concentration of the antioxidants present in the tested sample. The principle of this method is based on the reduction of ferric-tripyridyltriazine complex (FeIII-TPTZ) to a ferrous (FeII) derivative with an intense blue color that absorbs at 593nm (Benzie & Strain, 1996).

$$Fe(TPTZ)_2^{3+} + ArOH \rightarrow Fe(TPTZ)_2^{2+} + ArO^{\bullet} + H^+$$

Figure 6. FRAP method reaction where ArOH = phenol (Apak *et al.*, 2016)

Even though FRAP assay has the aforementioned benefits, it suffers from certain drawbacks. FRAP and other assays that transfer electrons to reduce Fe(III) to Fe(II) would give erroneous results because being a reduction product Fe(II) could potentially generate reactive species such as hydroxyl radicals through Fenton-like reactions with H_2O_2 leading to a redox cycling of phenolics (Apak *et al.*, 2016).

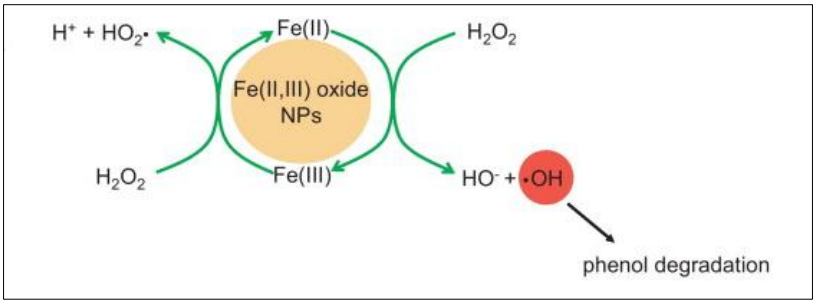

Figure 7. Fenton like Reaction (Rusevova *et al.*, 2012)

2. Trolox equivalent antioxidant capacity assay (TEAC/ABTS•+)

The trolox equivalent antioxidant capacity (TEAC) assay uses intensely colored cation radicals of ABTS•+ to accept hydrogens or electrons from antioxidant molecules (Apak *et al.*, 2016). The blue/green ABTS•+ radical cation is reduced by both lipophilic and hydrophilic antioxidants and the concomitant decolorization of this cation that takes place is monitored at 734 nm (Floegel *et al.*, 2010).

The antioxidant capacity is measured as the capability of the antioxidant present in the tested sample to reduce the color of the ABTS•+ through intercepting initial oxidation and avoiding the production of ABTS•+ or by reacting directly with the radical cation (Apak *et al.*, 2016). Based on these two approaches, two versions of this assay are possible. The first version gives inaccurate results from overestimation of antioxidant activity. The second version which uses potassium per-sulfate as an oxidizing agent produces ABTS•+ which is then reacted with the antioxidants and the accompanying decrease in the color intensity is monitored at 734 nm (Figure 8). The results are reported as trolox equivalent (Schaich *et al.*, 2015).

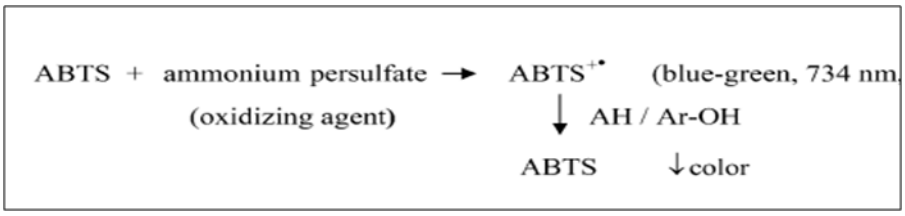

Figure 8. TEAC assay reaction (Schaich *et al.*, 2015)

The main drawbacks of the ABTS assay are that the reaction is not over at the usually used time of 6 min and the free radical being used is not present in vivo.

E. Determination of polyphenol intakes

Determination of food intakes of populations differ in the method used to measure individuals diets with some utilizing the twenty four hour diet recalls (24 HR) and others using food frequency questionnaire (FFQ). The other used methods include forty-eight hour recall (48hR), a Diet History Questionnaire (DHQ), National Survey Questionnaire and Food Balance sheets (FBS). The 24 HR provides detailed information on all consumed foods and beverages during the 24 hours of a previous day while the food frequency questionnaire FFQ gives information on how often a food is consumed over a long period of time, normally, the previous year in addition to the portion size. FFQ method is prone to large random errors and needs validation before usage. Even though 24 HR recall is more accurate than FFQ, one single 24 HR does not represent the habitual intake so multiple recalls are needed in non-consecutive occasions. This is very important when dealing with PP intake due to the fact that some foods that are rich in PPs are consumed seasonally.

Moreover, DHQ is the most time consuming and expensive method. It is not suitable for large population since it requires collecting information on both quantity and frequency of the foods consumed over a selected period of time.

Further, the FBS is an inadequate method for assessing PP intake because it may not cover all food items and often provides a gross estimate rather than representing the actual consumption. FBS is usually used to collect information on a country's food supply showing the amount of foods that are potentially available over a reference period (Pinto and Santos 2017).

A number of epidemiological studies and clinical trials have determined the antioxidant capacity AC of selected plant foods and polyphenol intake for different populations. The estimation of PP intake and capacity is the first step in assessing the protective effects of PP against the risk of chronic disease. Moreover, assessing dietary intake and behaviors of subpopulation groups is fundamental to establish national food

and health guidelines to sustain productivity and health (Pinto and Santos, 2017; Torres and Farah, 2017).

Studies assessing antioxidants in fruits, vegetables and beverages can be grouped in three categories:

1) Those estimating the dietary intake of polyphenol through measuring PP content: in Japan (Fukushima *et al.*, 2009; Fukushima *et al.*, 2014; Sakakibara *et al.*, 2003), Poland (Cieślik *et al.*, 2004), France (Brat *et al.*, 2006) and in USA (Vinson *et al.*, 2001).

2) Those evaluating antioxidant capacity; in Brazil (Torres *et al.*, 2017), Spain (Alonso *et al.*, 2004) and in Italy (Pellegrini *et al.*, 2003).

3) Studies assessing both phenol content and capacity; in Poland (Zujko & Witkowska, 2011), USA (Chun *et al.*, 2005), Chile (Álvarez *et al.*, 2016), Japan (Takebayashi *et al.*, 2013), Australia (Lako et *al*,. 2017) and Czech Republic (Stratil *et al.*, 2006).

However, with few exceptions, these studies analyzed only the extractable polyphenols and, accordingly, underestimate the PP intake due to the lack of data on the NEPP. NEPP content was assessed in three fruits (apples, peach and nectarines) in Spain (Arranz *et al.*, 2009) and in different kinds of fruits and vegetables along with their antioxidant capacity in four European countries (France, Germany, Netherlands and Spain) (Pérez-Jiménez *et al.*, 2005).

No data exists on antioxidant capacity of the Lebanese diet and total antioxidant intake of the Lebanese population. Therefore, the purpose of this study was to evaluate the relative contribution of the most commonly consumed fruits, vegetables and beverages in Lebanon to the AC of the population's diet.

CHAPTER III

MATERIALS AND METHODS

A. Study design

The food consumption data used in the present study was obtained from a National survey conducted in Lebanon between May 2008 and April 2009 (Chamieh *et al.,* 2015). The study sample was based on the sampling frame provided by the National Survey of Household Living Conditions, which was conducted by the Ministry of Social Affairs/ Central Administration of Statistics in collaboration with the United Nations Development Program (UNDP), and which covered primary residences across the Lebanese territory. The sample was drawn from randomly selected houses based on stratified cluster sampling. The strata were the Governorates of Lebanon comprising the totally urban capital Beirut and five other governorates (Mount Lebanon, North, Beqaa, Nabatiye, and the South) that are a mixture of rural villages and urban cities. Healthy Subjects (1244 men and 1453 women) were randomly chosen including those ≥ 20 years old and excluding pregnant, lactating women and those with mental disabilities. Food consumption data was obtained by a 24 hour recall that was conducted by trained nutritionists using face-to-face interviews and following the 5-step multiple pass method for more accuracy (Chamieh *et al.,* 2015).

B. Preparation and selection of food samples

Food samples were selected according to the following strategy: the consumption data of individual items were listed in decreasing order until at least 85% of the consumption in each food group (beverages, fruits and vegetables) was covered (Appendix X). The varieties selected for each fruit and vegetable were based on horticulturist while the beverages were chosen according to the most sold brands based on informal feedback from supermarkets and consumers (Table 1a, 1b).

Table 1(a). Brands of the selected beverages items

Beverages
Black tea Ahmad/ Horse Head/ Lipton
Coffee Turkish Maatouk/ Najjar/ Daniel café
Instant Coffee RedMug/ Matinal/Gold
Pineapple juice Tropicana (Pineapple drink, 10% pineapple juice concentrate) Maccaw (100% pineapple juice from concentrate) Xtra (Pineapple drink from concentrate: juice concentration 30% minimum)

Table 1(b). Varieties of the selected fruits and Vegetables

Vegetables	*Fruits*
Tomato Green House/ Cherry Tomato	**Apples** Granny smith/ Red Delicious /Golden
Cucumber Lebanese cucumber	**Banana** Baby banana/Cavendish/ Grand Nain
	Clementine Lebanese
	Oranges Navel oranges/ Common oranges
	Pear (Lebanese)
	Olives Baladi, black/ Baladi, green/ Ayroni, black/ Green, north
	Grapes Helwani –red/ Beitamouni- white/ Maghdoushi

	Peach Nectarine/ Babcock yellow
	Apricot Um Hussein/ Ajami
	Water melon Crimson-oblong
	Cantaloupe Ananas

Four beverages (Black tea, Turkish coffee, instant coffee and canned pineapple juice), 2 vegetables (Cucumber and tomato) and 11 fruits (Banana, apples, oranges, pears, clementine, grapes, watermelon, cantaloupe, peaches, apricots and olives) were analyzed in this study. The fresh fruits and vegetables were purchased from local supermarkets in Beirut, Lebanon and were washed with tap water, dried, chopped, frozen and freeze dried (LABCONCO FreeZone 6 freeze dryer, 8811 Prospect Ave, Kansas City, MO 64132, USA). The stones were removed from olives, peach and apricot prior to analysis. The peels were discarded and only the edible portions from bananas, oranges clementine, water melons and cantaloupe were analyzed.

The edible portions were weighed before and after freeze drying to calculate the moisture content. The dried samples were ground to powder using a "Moulinex Mill grinder" to a particle size of < 0.5 mm and then stored at ≤ 4 ^{o}C for no more than one month until analyzed.

For the preparation of black tea, 2g of tea leaves were added to 200 ml boiling distilled water and left for 5 min. The Turkish coffee was prepared by adding 5g of coffee powder to 90 ml of distilled water. A slow heating until the boiling was performed to allow for the development of foam on the beverage surface. Then, the process is interrupted for few seconds before repeating the boiling to facilitate the precipitation of insoluble compounds (Severini *et al.,* 2017). Instant coffee was prepared by adding 5g of coffee to 200 ml boiled distilled water. The coffees were filtered using a Whatman n^{o} 1 paper before analysis.

C. Chemicals

All chemicals used were of analytical grade. Gallic acid, sodium carbonate (Na_2CO_3), Folin-Ciocalteu, methanol, acetone, sodium acetate trihydrate, glacial acetic acid, concentrated HCl, TPTZ (2,4,6-Tri(2-pyridyl)-s-triazine), $FeCl_3$ (Iron Ⅲ chloride), ABTS (2,2'-azino-bis(3-ethylbenzthiazoline-6-sulphonic acid), potassium persulfate, butanol, sulfuric acid (H_2SO_4), ethanolamine, ammonium acetate were purchased from Sigma-Aldrich (Taufkirchen, Germany).

D. Isolation and determination of extractable polyphenols

For the isolation of extractable polyphenols from the solid samples, the ground food sample (~ 1g) was placed in a capped centrifuge tube and acidic methanol/water/HCL (20 mL; 50:50 v/v, pH 2) was added and the tube was thoroughly shaken at room temperature for 1 h. The tube was then centrifuged at 3500 rpm (Thermo Electron corporation IEC Cl3R centrifuge, USA, 81 Wyman Street Waltham, MA 02454) for 10 min and the supernatant was recovered. Acetone/water (20 mL, 70:30 v/v) was added to the residue and the shaking and centrifugation was repeated. The methanolic and acetonic extracts were combined to yield the extractable polyphenol fraction. Extractions were performed in triplicates. The residues of these extractions were used for the analysis of the non-extractable polyphenol fractions (NEPA) (Arranz *et al.,* 2009).

For tea, coffee and instant coffee extraction, the sample (5 mL) was added to acetone/ water (25ml, 7:3 v/v) solution. Pineapple juice (10 g) was stirred with acetone/ water (10ml, 7:3 v/v) solution for 30 min. The mixture was passed through Whatman n^o 1 filter paper and the filtrate collected to correspond to the raw extract (RE). Distilled water was added to the RE to reduce the proportion of acetone to 7% and the diluted RE (2ml) was loaded onto an Oasis HLB cartridge 1cc (30mg; from Waters) to remove any interfering water-soluble components. The cartridge was eluted with 2×2 ml distilled water and the recovered volumes of the washing extract (WE) were measured. Prior to use, the cartridges were conditioned by washing with 4ml methanol followed by 2×4ml distilled water (George *et al.,* 2005).

E. Samples extraction (Non-Extractable polyphenols)

The residues left after isolation of the extractable polyphenols were freed from solvents freeze dried and stored at 4^{o} C until used.

1.Non extractable Proanthocyanidins (NEPA)

The residue (50 mg) was treated with butanol/concentrated HCl (10 mL; 95:5 v/v) and $FeCl_3$ (0-7 g) at 100 ^{o}C for 60 min. The tubes were then centrifuged (Thermo Electron corporation IEC Cl3R centrifuge, Milford, MA 01757) at 3800 rpm for 10 min and the supernatant was collected. After two washings with 5 ml butanol and centrifugation, the supernatants were combined and the absorbance was measured at both 450 and 555 nm (Arranz *et al.,* 2009).

The PA concentrations (mg/L) were determined from the sum of absorbance at 450 and 555 nm expressed as mg PA per 100 g of dry weight. The standard curve was developed using a carob concentrate (20mg) prepared under the same conditions as the sample (Appendix ΙΙ).

F. Determination of phenol content

The total phenol content was determined by the Folin-Ciocalteu procedure according to ISO 14502-1:2005. The sample (1 mL; diluted when necessary) was mixed with the Folin-Ciocalteu reagent (5 mL; 10% volume fraction) and left at room temperature for 5 min. Sodium carbonate solution (4 mL; 7.5% mass concentration) was added to each tube. The tubes were allowed to stand at room temperature for 60 min and their absorbance measured at 765 nm (Thermo Scientific Evolution 300 UV-VIS, Waltham, MA, USA). The total phenolic content was expressed as mg Gallic acid equivalents (GAE) / 100 g fresh weight and ml for solid and liquid samples, respectively. A standard curve was prepared from Gallic acid solutions (0-60 μg/ml) (Appendix ΙΙΙ).

G. Antioxidant Capacity Assays

1. Ferric-Reducing Antioxidant Power (FRAP)

The antioxidant capacity of the food samples were determined by the FRAP assay according to the method of Benzie and Strain (1996) with some modifications. The FRAP reagent was prepared using 10:1:1 acetate buffer (300 mM solution): TPTZ (2,4,6-tripyridyl-striazine) (10 mM solution): $FeCl_3.6H_2O$ (20 mM solution). The solution was prepared daily before each analysis. The sample (100 µL or a dilution thereof)) was mixed with distilled water (900 µL) and the FRAP reagent (2mL). The tubes were mixed well and then stored in the dark for 30 min and the absorbance measured at 593nm. The antioxidant capacity of the tested samples was expressed as mmol Fe (II) / L or Kg fresh weight. The standard curve was prepared from ferrous sulfate solutions (0-100 µM) (Appendix Ⅳ).

2. Trolox equivalent antioxidant capacity assay (TEAC/ABTS•+)

The antioxidant capacity of the samples was also measured by the Trolox equivalent antioxidant capacity assay according to Floegel *et al.* (2010) with some modifications. The $ABTS^{\bullet+}$ solution was prepared from 7 mM ABTS and 2.45 mM potassium persulfate in a volume ratio of 1:1, and then incubated in the dark for 15-16 h at room temperature. The $ABTS^{\bullet+}$ working solution was prepared by diluting the stock solution with methanol to an absorbance of 0.70 ± 0.05 at 734 nm. The sample (100 µL) was mixed with 3.8 ml $ABTS^{\bullet+}$ working solution, incubated in the dark for 30 min and the absorbance of the mixture measured at 734 nm. Trolox was used as a reference standard (Figure 4), and the results were expressed as µmol Trolox/g fresh weight for fruits and vegetables and mmol Trolox/L for beverages. The standard curve relating absorbance to concentration of Trolox (0 – 500 µM) is presented in (Appendix V).

CHAPTER IV

RESULTS AND DISCUSSION

A. Total phenol contents of beverages, fruits and vegetables

1.Beverages

The phenol contents of the analyzed beverages are presented in Table 2. Instant Coffee (NESCAFÉ, Nestlé Middle East) contained the highest amount of polyphenols (375 mg/100ml) followed by Turkish coffee (369.8 mg/100ml), Black tea (98.8 mg/100ml) and pineapple canned Juice (11.2 mg/100ml) (Figure 9).

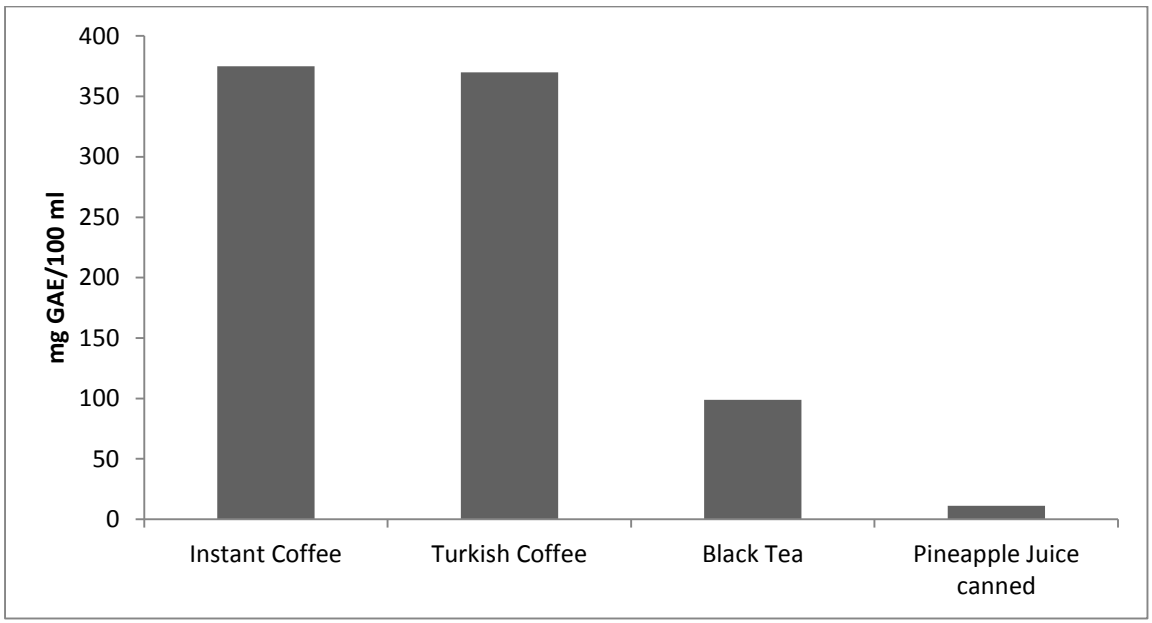

Figure 9.Total phenol content in the most consumed beverages in Lebanon

According to the National survey, the average consumption of beverages was 0.28 L/day per individual. Black Tea was the most consumed beverage estimated by 153.9 ml/day and 55.5% of the total consumption. Turkish coffee was the second mostly consumed beverage with consumption 32.51 mL/day and 11.7% of the total beverages consumption. Moreover, Black tea was the largest source of total polyphenol consumption from beverages providing the Lebanese individual by 152.2 mg of PP per

day. The Second and third largest source were Turkish coffee and Instant Coffee providing 120.2 and 99.41 mg PP/ day respectively, however, pineapple canned juice was considered the lowest source with 3.02 mg PP/ day.

Table.2 Dietary intakes of total phenols from the most consumed beverages in Lebanon

	Average intake mL/day/person		**Total phenols[1] (mg GAE/100 mL)**	**Total phenols intake mg/day/person**
Beverages		**(%)**[a]		
Black Tea[2] (n = 3)	153.9	55.5%	98.8 ±4.22 (94.26-102.5)	152.2
Turkish Coffee[3] (n = 3)	32.51	11.7%	369.8 ±5.6 (365-376)	120.2
Instant Coffee[4] (n = 3)	26.51	9.55%	375±52.9 (315-415)	99.41
Pineapple Juice canned (n = 3)	27.01	9.73%	11.2 ±5.13 (6.37-16.6)	3.02
Total	239.9		854.8	374.83

n= number of Brands analyzed

1. Averages of triplicate determinations of 3 different brands; Mean ± SD; values in brackets are ranges

2. Prepared by adding 200 mL hot water to 2g coffee and steeping for 4 minutes

3. Prepared by adding 90 mL hot water to 5g grounded coffee and boiling for couple of minutes

4. Prepared by adding 200 mL hot water to 5 g instant coffee granules

[a] Percentage of consumption within Beverages group

The total phenol content of black tea ranged between 94.26 and 102.5 mg /100 mL was similar to that reported for different brands of black tea from (Lebanon) at 80.5-134.9 mg gallic acid equivalent (GAE)/g (Khokhar *et al*., 2002), UK at 83.2 mg GAE/g /g (Lomillo, 2011) and in the Phenol-Explorer database (104 mg/100ml) (Pérez-Jiménez *et al.,* 2010). Furthermore, the values were higher than that in analyzed black tea in Poland at 72 mg GAE /100mL (Zujko & Witkowska, 2014).

Of the total world tea production, 76-78% is produced as black tea which ranks highest in worldwide consumption of beverages (Fernando& Soysa, 2015).The main polyphenolic compounds in tea are the flavan-3-ols (catchenins) including (−)-epicatechin (EC), (−)-epigallocatechin (EGC), (−)-epicatechin gallate (ECG), (−)-epigallocatechingallate (EGCG), (−)-gallocatechins (GC) and (−)-gallocatechin gallate (GCG). During the fermentation process catechins are oxidized to theaflavins and thearubigins which are responsible for the color change from colorless to the color of the Black tea (Fernando& Soysa, 2015).

The TP contents of Turkish coffee (365 - 376 mg GAE/ 100 mL) were within the bracket (306.8 mg GAE/100mL - 400 mg GAE/100 mL) reported for 12 brands of Turkish coffee ground from medium roasted coffee beans (Gorjanović *et al*., 2017). However, our values were higher than those of Turkish coffee brews with different roasting degrees with values registered at 337.23 mg GAE /100mL, 317.88 mg Gallic acid/100 ml and 220.74 mg/100ml for light roasted, medium roasted and dark roasted brews, respectively (Nakilcioglu-Tas, 2018).

The coffee beans available in the market are produced from either *Arabica* or *Robusta* species or a blend of both in different proportions (Al-Ghafari & Althaqafi, 2017). The *Arabica* variety is preferred in the production of Lebanese Coffee, also known as Turkish coffee, for its milder and flavorful taste (Mikhael, 2017; Nakilcioglu-Tas, 2018).

Hydroxycinnamic acids and Hydroxybenzoic acids are the major groups of polyphenols in coffee with the major phenolic acids present being ferulic acid, p-coumaric acid, caffeic acid, vanillic acid, syringic acid and chlorogenic acids (Somporn *et al*., 2012). During roasting, phenolic compounds such as chlorogenic acid partially degrade with some forming polymeric compounds through the Maillard reaction thereby

causing a decrease in the phenol content of coffee and the aforementioned differences in TPC between light, moderate and dark roasted beans. On the other hand, the antioxidant capacity would remain unchanged or increased by the Maillard reaction-formed melanoidins which possess high antioxidant activity (Yilmaz *et al*., 2014).

In the industry, roasted and ground coffee beans are often subjected to agglomeration which consists of boiling, steeping, filtration and application of pressure to extract the water-soluble compounds marketed as instant coffee (Al-Ghafari & Althaqafi, 2017). The analyzed instant coffee samples had a TPC ranging between 315-415 mg GAE/100 mL which is higher than that done by (Al Doghaither *et al.*, 2017) and (Lomillo, 2011) showing that total phenol content in Red mug Nescafé was 74.1 mg Gallic acid/ 100mL of 2 g of coffee and in Regular Nescafé ±94.4 mg Gallic acid/100mL coffee respectively. The instant coffee samples were prepared by adding 2g of coffee to 200ml and 250 ml water, while our samples were prepared by mixing 5g in 200ml water. Therefore, the difference in TPC results could be linked to the dissimilar way of preparation. Furthermore, instant coffee brews in Turkey showed higher results where phenol content ranged between 915.9-961.4 mg GAE/ 100mL. These samples were made up by pouring 200 ml water on 6g of coffee (Gorjanović *et al.,* 2017).

The lowest TPC amongst the surveyed beverages was that of canned pineapple juice with a mean of 11 mg GAE/100mL (Table 1). Processing of fruit juices is often accompanied by a decrease in their TPC (Mahdavi *et al*., 2010). The TPC of 13 samples of commercial pineapple juice consumed in Spain ranged between 17.3-48.1 mg GAE/ 100mL (Pastoriza *et al*., 2017), while for those available in the local stores in Tabriz-Iran showed a value of 35.74 mg GAE/100mL Mahdavi *et al.,* 2010). Moreover, ±20 mg GAE/100mL was detected in both commercial and pasteurized pineapple juice in Thailand (Abdullakasim *et al.,* 2007). It is noteworthy that in these studies the interfering vitamin C and sugars were not removed and might have contributed to the relatively-high TPC. This is corroborated by the mean TPC of 36 mg GAE/ 100 mL of canned pineapple juice obtained for samples without removal of the interfering vitamin C and sugars. Moreover, the TP contents didn't differ much between the different analyzed brands, even though they differ in the percentages of their juice concentrate contents, thereby suggesting mislabeling of the products.

Phenol content in most consumed Fruits and vegetables in Lebanon is present in the below graph (Figure10) and in (Table 3).

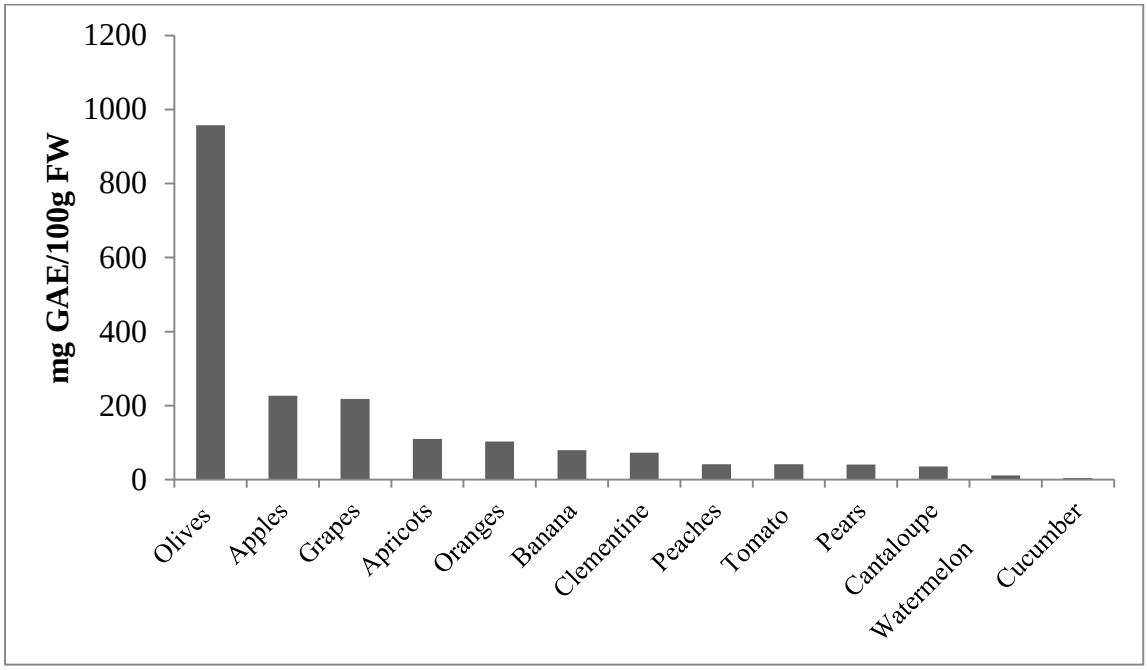

Figure.10 Total phenol content in fruits and vegetables

Results are on wet basis from triplicate of analyses and triplicate of food samples

Table .3 Total polyphenol content in the most consumed fruits and vegetables in Lebanon

	Consumption		**Average total phenol Content [a]**	
	g/day [b]	**(%)[c]**	**mg/100 g FW**	**mg/100 g DW**
Fruits				
Apples(n=3)	31.73	20.68	226.8 ±119.5(157-365)	1214.5 ±189(1056-1424)
Grapes(n=3)	23.1	15.06	217.8 ±56.3(183.8-282.9)	1508.4 ±135(1351-1589)
Banana(n=3)	15.62	10.18	80 ±60.6(32.3-148.3)	548 ±436(130-1000)
Watermelon(n=1)	12.52	8.16	11.3	119.15
Pears(n=1)	12.19	7.95	40.4	270.4
Peach(n=2)	9.67	6.3	41.4 ±36.9(38.8-44)	303.04 ±36.4(277-

				328.8)
Orange(n=2)	7.257	4.73	102.6 ±4.6(99.5-106)	766.15 ±7(761-771)
Olives (n=4)	6.66	4.34	957.9 ±597(449-1733)	1951.8±1341(1023-3911)
Cantaloupe(n=1)	4.62	3.01	35.7	333.5
Apricot(n=2)	4.4	2.87	109.9±108(33.5-186)	813.8±892(182-1445)
Clementine(n=1)	3.72	2.42	72.5	640
Total	131.48		1896.3	7920.7
Vegetables	**g/day [b]**	**(%)[c]**	**mg/100 g FW**	**mg/100 g DW**
Tomato(n=2)	29.66	47.1	41.3±19.4 (27-55)	483.2±182(354-612)
Cucumber(n=1)	24.91	39.6	4.39	268.75
Total	54.57		45.69	751.95

n= number of varieties analyzed

[a] The Total phenol content expressed in fresh and dry weight of the sample

[b] Amount of daily consumption expressed in grams per individual

[c] Percentage of consumption within fruits and vegetables group

2.Fruits

Fruits constitute the largest part of agricultural production in Lebanon at 26%. Citrus, apples, grapes and bananas are the main commodities within this category. Apples account for 23% of total fruit production followed by banana at 18% and grapes at 14% (IDAL 2015). Furthermore, apples were the most consumed fruit by the polled subjects (Chamieh *et al.*, 2015) accounting for 20.68% of all consumed fruits, followed by grapes and bananas at 15.06% and 10.18%, respectively.

In the present work, apples were the largest source of total polyphenol consumption from fruits (72 mg GAE) followed by olives (63.8 mg GAE) and grapes (50.3 mg GAE) (appendix VI).

- Apple

Apples are one of the main dietary sources of phenolic antioxidants. The phenolic content vary among different varieties, cultural practices, growing conditions, degree of

ripeness and storage conditions (Henriquez *et al*., 2010). The mean phenolic content of apples consumed in Lebanon was 226.8 mg Gallic aid/100g FW with the highest levels being registered for apples with red skin (var. Red Delicious) at 337.5 mg/100g FW as compared to 157 mg/100g FW and 158 mg/100g FW for green (var. Granny Smith) and yellow-skinned (var. Golden Delicious) apples, respectively (Table 2). The total phenol content of Red Delicious apples (337.5 mg/100g FW) was slightly higher than that of the same variety (250 mg/100g FW) while TPC of Granny Smith (157 mg/100g FW) were slightly lower than those reported for the same variety (200 mg/100g FW) grown in Chile (Henriquez *et al*., 2010). Moreover, Golden Delicious apples had a relatively-higher phenol content at 158 mg/100g FW than that reported for the same variety (126.8 mg/100g FW) grown in Poland (Podsędek *et al*., 2014). In general, the mean average of TPC in Lebanese apples (226.8 mg/100g FW) was higher than those reported for apples by other workers at 128.29 mg/100g FW (Pinto & Santos, 2017), 85 mg/100g FW (Takebayashi *et al*., 2013), 179.1 mg/100g FW (Brat *et al*., 2006) and 118.3 mg/100g FW (Chun *et al*., 2005). These differences could be attributed to the different apple varieties studied, cultural and growing practices.

Phenolic compounds found in apples belong to a number of groups: 1) hydroxybenzoic acids including *p*-hydroxybenzoic acid, protocatechuic acid, gallic acid, syringic acid and gentisic acid, 2) hydroxycinnamic acids and their derivatives: *p*-coumaric acid, caffeic acid, ferulic acid and chlorogenic acid, 3) glycosylated flavonols (mainly quercetin), 4) dihydrochalcones mainly phloridzin and its derivatives, 5) anthocyanids, 6) monomeric flavanols notably epicatechin and catechin, and 7) oligomeric flavanols of the procyanidin type. Phenolic compounds in plants rarely exist in free form and are normally present in the form of glycosides or esterified with carboxylic acids. However, among edible fruits, apples have the highest amounts of free phenolics that are accessible for immediate absorption into the blood stream. Additionally, the phenolic composition differs between the peel and flesh of apples fruit: In general, apple peels have higher total phenol compounds, total procyanidins and flavonoids than the flesh, even though the flesh of certain varieties is richer in certain individual phenolic compounds compared to the peel. ‘‘Red Delicious’ is among the varieties that have more chlorogenic acid in the peel than in flesh in contrast to the pattern in ‘Golden Delicious ’and ‘Granny Smith’ (Kalinowska *et al.*, 2014).

- Grapes

The phenolic compounds in grapes are spread in the leaf and stem of the vine and, chiefly, in the skin and seeds of the fruit with very little being present in the pressed juice. Grapes are consumed as fresh fruit, raisins, juice or in the form of wine. The difference in soil composition, geographic region and cultivation practices leads to differences in the total phenol content of grape skins. Phenolic compounds in grapes include anthocyanins which are responsible for pigment and mainly exist in the grape skin, proanthocyanidins (in seeds and skin), flavanols, flavonols, stilbenes (mainly resveratrol) and phenolic acids. Grapes also contain flavonoids such as catechins, epicatechin and procyanidin polymers. Anthocyanins are the main polyphenols in red grapes, while flavan-3-ols are more abundant in white varieties (Xia *et al*., 2010).

The mean total phenol content of the analyzed white- and red-skinned grapes was 217.8 mg/100 g FW (1508.4 mg/100g DW). These values were slightly higher than reported for varieties from France at 195 mg/100g FW (Brat *et al*., 2006) and slightly lower than reported for grapes consumed in Europe at 1870 mg/10g DW (Pérez-Jiménez & Saura-Calixto, 2015). Grapes with white skin had TPC of 1351 mg/100g DW which is much higher than white grapes from the Polish market (771-816 mg/100g DW; Cieślik, *et al*., 2006) and the USA (83.59 mg/100g FW; Chun *et al*., 2005). It is noteworthy; that the higher values obtained in the present could be due, at least in part, to the inclusion of the skin and seeds in the analyzed samples in contrast to other studies which did not include the seeds and skins in the analysis (Brat *et al.,* 2006; Chun *et al*., 2005).

- Banana

The mean total phenol content of banana fruit was 80 mg/100g FW with the highest TPC being registered for the Grand Nain variety (148 mg/100g FW) and the lowest for the Cavendish type (32.35 mg/100g FW) which was comparatively lower for the Cavendish variety consumed in France (51.5 mg/100g FW; Brat et al., 2006). The TPC of 8 varieties of bananas from Malaysia ranged within 13-36 mg/100g FW (Sulaiman *et al.,* 2011) and the reported mean TPC for bananas, unknown varieties, from USA was 112.79 mg/100g FW (Chun *et al.,* 2005), Poland 104 mg/100g FW (Zujko *et al*., 2012) and Japan 62 mg/100g FW (Takebayashi *et al.,* 2013). Further, the TPC in three groups

of banana consumed in China was 57.13 mg/ 100g FW for general bananas, 29.07 mg/ 100g FW for cooking bananas and 25.55 mg/ 100g FW for royal bananas (Fu *et al.*, 2011).

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Bananas contain varied phenolic antioxidants including catechin, epicatechin, tannins, and anthocyanins in addition to phenolic acids as ferulic, sinapic, salicylic, gallic, *p*-hydroxybenzoic, vanillic, syringic, gentisic, and p-coumaric acids. Moreover, the flavonoids present in this fruit include quercetin, myricetin, kaempferol, and cyanidin (Sidhu & Zafar, 2018).

- Watermelon

Watermelons contain different types of antioxidants including carotenoids which impart to the flesh its characteristic red color (lycopene and β-carotene), vitamins (A, B, C and E) and phenols (Choudhary *et al*., 2015). The TPC in watermelon was 11.3 mg/100 FW (119.15 mg/100g DW) similar to those reported by different workers (Pierre Brat 11.6 mg/100g FW, Brat *et al*., 2006; 14 mg/100g FW, Takebayashi *et al*., 2013; 112 mg/100g DW; Pérez-Jiménez & Saura-Calixto, 2015).

- Pear

The Pear fruit is consumed worldwide in the fresh and processed (e.g drink, candy, preserved fruit and jams) forms. The pear contains varied phenolics including arbutin, chlorogenic acid, catechin, quercetin, kaempferol, various

hydroxycinnamoylmalic acids and their ethyl esters, hyroxycinnamoyl malates, and procyanidins and triterpenes which are found predominantly in the peel (Li *et al*., 2014). The mean TPC of pear fruits analyzed in the present work was 40.4 mg/100g FW which was slightly lower than those reported in the literature 68 mg/100g FW for Australian pears (Fu *et al.,* 2011), 69.2 mg/100g FW for pears from France (Brat *et al.,* 2006), 72 mg/100g FW for Polish pears (Zujko & Witkowska, 2014) and listed by the Phenol-Explorer database at 59.6 mg/100g FW (Pinto & Santos, 2017). However, results were higher than Japanese pear 12 mg/100g FW and Chinese pear (var. Royal) 34.84 mg/100g FW (Takebayashi *et al.,* 2013; Fu *et al.,* 2011).

- Peach

The mean TPC of the analyzed peach fruits was 41.4 mg/100g FW with the TPC of Nectarine (white flesh) being higher at 44 mg/100g FW than yellow-fleshed Babcock variety at 38.82 mg/100g FW. These results were similar to the TPC determined for 11 peach cultivars (white, yellow and nectarine) ranging between 14-50 mg/100g FW (Tavarini *et al.,* 2008) and 15 different genotypes with mean of 36.4 mg/100g FW (Cantín *et al.,* 2009). The phenolic contents of nectarines available in the Lebanese and Spanish markets were similar at 44 mg/100g FW and 45.8 mg/100g FW (Arranz *et al.,* 2009), respectively. Peaches are rich in vitamin C, carotenoids and phenolic compounds predominantly anthocyannins including cyanidin-3-glucoside and cyanidin-3-rutinoside (Cantín *et al.,* 2009).

- Orange and Clementine

Citrus fruits account for 17% of total fruit production in Lebanon (IDAL 2015). Major phenolic compounds present in citrus fruits include hydroxycinnamic acids (ferulic, *p*-coumaric, sinapic and caffeic acids) and flavonoids where flavanones are the most prevalent and present mainly as glycosides (hesperidin and narirutin). Hesperidin is claimed to have therapeutic properties including anti-inflammatory, antihypertensive, diuretic, analgesic, and hypolipidemic activities (Park *et al.,* 2014).

The analyzed oranges showed similar TPC in Navel oranges at 99.48 mg/100g FW and the common oranges at 105.8 mg/100 FW (Table 2). The mean TPC of the oranges at 102.6 mg/100g FW (7661.5 mg/100g DW) was similar to that reported at 112.29

mg/100g FW (786.5 mg/100g DW) (Pérez-Jiménez & Saura-Calixto, 2015; Chun *et al.*, 2005). The Navel oranges showed higher TPC (99.48 mg/100g FW) as compared to Navel orange from China (51.09 mg/100g FW) and USA (61.08 mg/100g FW) (Fu *et al.,* 2011). Likewise, the common oranges also showed higher phenolic content when compared to those from France (31 mg/100g FW; Brat *et al.,* 2006).

Clementine, a hybrid between orange and mandarin, is also considered within the citrus fruits. Clementine exhibited a TPC of 72.5 mg/100g FW (640 mg/100g DW) which is higher than that from France 30.6mg/100g FW (Brat *et al.,* 2006) and lower than that of Algerian Clementine Cultivars ranging between 3108.7- 4046.2 mg/100g DW (Boudries *et al.,* 2012).

- Olives

Olives are the second largest source of total polyphenols in the fruit group according to the national survey (Table 4). Olive is a major crop in Lebanon covering around 21% of the total cultivated area. Olives make up 4% of the total agricultural production with 80% being processed to olive oil and the remaining 20% is consumed as table olives (IDAL 2015). Even though, olive production increased from 1990 to 2007 the quantities produced are still not sufficient to meet the national demands (Chehade *et al.,* 2015). Olive trees are planted chiefly in the northern and southern parts of the Lebanon. The olive varieties analyzed in the present work are the most common cultivated varieties/accessions in Lebanon. Both black and green olives were analyzed for total phenols with mean TPC being 1382.5 mg/100g and 477.8 mg/100g FW, respectively.

Table.4 Total Phenol Content of Olives Expressed in Fresh and Dry Weight

Olives Variety	TPC mg/100g FW	TPC mg/100g DW
Black Olives (n =2)	1382.5 ±495	2815.95 ±1579.9
Green Olives (n =2)	477.8 ±71.4	1087.79 ±90.4
Mean TPC	930.15	1951.87

The Baladi and Ayroni black-type olives showed slightly lower TPC (2815.95 mg/100g DW) than black olives from Greece 3401 mg/100g DW (Tsantili, 2014) and higher than that reported for the same type at 569.2 mg/100g FW (Pinto & Santos,

2017). Lower TPC were reported for olives in the Chinese market (21.68 mg/100g FW; Fu *et al*., 2011), and olives with different physical appearance (color) ranging between 20-590 mg/100g DW (McDonald *et al.*, 2001).

The major classes of phenolic compounds in the olive fruits include phenolic acids (main hydroxycinnamic acid derivative is verbascoside), phenolic alcohols (3,4-dihydroxyphenylethanol (hydroxytyrosol or h-tyrosol) and *p*-hydroxyphenylethanol (tyrosol), flavonoids (luteolin 7-O-glucoside, rutin and apigenin 7-O-glucoside, and the anthocyanins, cyanidin 3-O-glucoside and cyanidin 3-O-rutinoside) and secoiridoids (oleuropein and ligstroside). The phenols vary in amount depending on the variety and degree of ripening. Oleuropein, is the component responsible for bitterness in olives and progressively decreases as the fruit ripens and accumulates phenolic compounds most notably demethyloleuropein and 3, 4-dihydroxyphenylethanol (Arslan, & Özcan, 2011).

Generally, olives are not consumed in raw form due to their excessively astringent and bitter taste. Accordingly, olives are cured to get rid of the undesirable tastes and develop the flavor and texture prized by the consumers. Table olives differ from raw olives qualitatively and quantitatively in their phenolic composition. The phenols and other water soluble components can diffuse from the olive fruits to the surrounding medium (brine) and vice versa and other concomitant changes take place. The glycosides formed (oleoside 11-methyl ester, hydroxytyrosol-1-O-rhamnosylglucoside) are hydrolyzed to hydroxytyrosol during lactic fermentation in green olives. Further, acid hydrolysis of hydroxytyrosol and tyrosol glucosides, anthocyanins, luteolin 7-glucoside, oleuropein, verbascoside, ligstroside and some of the derived products (caffeoylrhamnoglucoside, oleuropein derivatives, ligstroside aglycone) also takes place during the fermentation of black olives in addition to the polymerization of anthocyanins (Boskou, 2005).

- Cantaloupe

Cantaloupes are widely consumed for their pleasant flavor and good nutritional value. The cantaloupe fruit is cultivated in a range of varieties including orange flesh cantaloupes, green flesh honeydew, and mixed melons. It is a good source of vitamin C, β-carotene and phenols (Ismail *et al.*, 2010). The cantaloupe fruit analyzed in the present work had TPC of 35.7 mg/100g FW (333.5 mg/100g DW) in agreement with

values reported by other workers (26.4 mg/100g FW, Ismail *et al.*, 2010; 21.13-22.68 mg/100g FW, Koh *et al.*, 2016; 31.5 mg/100g FW, Fu *et al.*, 2011; 28 mg/100g FW, Takebayashi *et al.*, 2013). However, the obtained value was slightly higher than cantaloupes consumed in Europe at 427.8 mg/100g DW (Pérez-Jiménez & Saura-Calixto, 2015).

- Apricot

Apricots are rich in vitamins A, B, and C as well as polyphenols. The phenolic compounds in apricots are varied and include catechin, epicatechin, *p*-coumaric acid, caffeic acid, ferulic acid and their esters and chiefly chlorogenic acid; the flavonols are found mostly as glycosides and rutinosides of quercetin (Sochor *et al.*, 2010). The mean TPC of apricots was 109.9 mg/100 g FW with big difference being noted between Um Hussein (198 mg/100g FW) and Ajami (33.56 mg/100g FW) apricots. The TPC of different genotypes of apricot have been reported to range between 41-170 mg GA/100g FW (Sochor et al., 2010), 20.78 -75.76 mg GAE 100 g FW (Leccese *et al.*, 2007); mean TPC for apricots have been reported at 179.8 mg GAE 100 g FW (Brat *et al.*, 2006) and 148 mg/100g FW for Polish apricots (Zujko *et al.*, 2012).

3.Vegetables

Tomatoes and cucumbers were the most consumed vegetables accounting for 86.7% of the total vegetables consumed in Lebanon as shown in (Table 3).

- Cucumber

The TPC in the most cultivated variety of cucumber was 4.39 mg/ 100 g fresh weight (Table 3) which is comparatively markedly lower than the values reported for varieties grown in Japan (15 mg/100g FW; Takebayashi *et al*., 2013), Poland (17 mg/100g FW; Zujko *et al*., 2012) and in Burkina Faso (21.7 mg/100g FW; Bayili *et al*., 2011). Moreover, our results expressed on dry mass of the fruit (2.68 mg /g DW) were also lower than those reported (10.6 mg/g DW; Stratil *et al*., 2006) and (18mg/g DW; Chandra *et al*., 2014). However, lower TPC levels (197.2 mg/100g DW) have been reported for cucumbers consumed in Europe (Pérez-Jiménez & Saura-Calixto, 2015) as

compared to the cucumbers analyzed in the present study (268.75 mg/100g DW, Table 3).

Cucumber is an important item in the Mediterranean diet since ancient times and is mainly consumed in its fresh state in salads or fermented to pickles. Cucumber is considered a rich source of phenolic compounds with several conjugated and glycosylated forms of caffeic, *p*-coumaric and ferulic acids being present (Abu Reidah, 2013). The relatively-low phenol content of the cucumber variety cultivated in Lebanon warrant the introduction of varieties with higher TPC into the country.

- Tomato

The mean TPC of the analyzed tomato varieties was 41.3 mg Gallic acid/ 100g FW with levels in cherry tomatoes (55.08 mg/100g FW) being double than those of the greenhouse tomatoes at 27.06 mg/100g FW (Table 2). The TPC levels in the analyzed cherry tomato (55.08 mg/100g FW) was similar to that grown in Southwestern Romania (55.78 mg/100g FW; Nour *et al*., 2013), lower than the Chinese cherry tomato (73.51 mg/100g FW; Fu *et al*., 2011) and markedly higher than that consumed in France (26.4 mg/100g FW; Brat *et al*., 2006). Moreover, the TPC of the green house tomato (27.06 mg/100g FW) was similar to those reported by some workers (23.69 mg/100g FW, Chun *et al*., 2005; 28 mg/100g FW, Takebayashi *et al*., 2013) and within levels of round red tomato varieties grown in Romania with TPC ranging between 19.7-33.7 mg/100g FW (Nour *et al*., 2013); however, it was slightly lower than that from India (35.9 mg/100g FW; Saxena *et al*., 2007).

Tomato (*Solanum lycopersicum*) is a widely cultivated and consumed commodity which is either consumed in the fresh state, as in salads, or in processed form as in ketchup, sauce, juice and puree. Tomatoes are rich in carotenoids mostly lycopene that makes up 80-90% of the total pigments in the ripe fruit in addition to α-, β-, γ- and δ-carotene, zeaxanthin and lutein. Moreover, tomatoes contain significant amounts of phenolic compounds including flavonols, quercetin, kaempferol and hydroxycinnamic acids mainly caffeic and chlorogenic acids. The tomato skin contains 98% of the total flavonols, as conjugated forms of quercetin and Kaempferol, thus accounting for the observed higher TPC in smaller tomatoes (cherry tomato) as compared to larger ones (green house) since smaller tomatoes have higher skin to volume ratio. In addition to

their higher TPC, smaller fruit (cherry type) have generally higher vitamin C content than their larger counterparts (Nour *et al*., 2013).

B. Antioxidant Capacity

The total antioxidant capacity (TAC) was measured using the ferric reducing antioxidant power (FRAP) and trolox equivalent antioxidant capacity (TEAC) assays (Table 4). The FRAP assay is based on the reduction of ferric-tripyridyltriazine complex (FeIII-TPTZ) to the intense blue color ferrous (FeII) derivative that absorbs at 593 nm (Benzie & Strain, 1996). The TEAC assay uses intensely colored cation radicals of ABTS•+ to accept hydrogen or electron from antioxidant molecules (Apak *et al.,* 2016). The blue/green ABTS•+ radical cation is reduced by both lipophilic and hydrophilic antioxidants and the concomitant de-colorization of this cation that takes place is monitored at 734 nm (Floegel *et al.,* 2010).

A high and positive correlation was obtained between the total phenol content of the analyzed food items and beverages and antioxidant capacity measured by FRAP assay ($r = 0.779$, $p < 0.01$) as well as the TEAC assay ($r = 0.867$, $p < 0.01$) (appendix VII). This indicates that phenolic compounds are the main components responsible for the reducing ability and free radical- scavenging capacity of the analyzed commodities.

Table .5 Total antioxidant capacities of the most consumed beverages, fruits and vegetables in Lebanon as measured by the FRAP and TEAC assays

	FRAP[1] (mmol Fe^{2+}/L or Kg FW[a])	TEAC[1] (mmol Trolox/L or Kg FW[a])
Beverages		
Instant Coffee[2] (n=3)	120.6 ±83.2 (32.5-198.2)	45.1 ±8.04 (39.6-54.3)
Turkish Coffee[3] (n=3)	14.6 ±0.35 (14.3-15)	14.35 ±0.42 (13.87-14.67)
Black Tea[4] (n=3)	13.6 ±6.3 (9.4-20.9)	6.82 ±1.1 (5.57-7.68)
Canned-pineapple	3.19 ±2.11 (1.81-5.63)	1.39 ±0.7 (0.89-2.2)

juice (n=3)		
Fruits		
Olives (n=4)	101.± 95.9 (47.3-244.5)	51.3± 17.1 (29.1-70.3)
Grapes (n=3)	29.6± 2.96 (26.5-32.4)	14.1± 4 (10.7-18.6)
Banana (n=3)	23.77±30.6 (3.5-59.03)	9.69± 9.19 (2.27-19.98)
Apricots (n=2)	21.28± 25.73 (3.08-39.48)	18.1±7.63 (12.7-23.5)
Apples (n=3)	14.6± 6.35 (10.78-21.9)	15.6± 8.7 (9.35-25.65)
Oranges (n=2)	7.266± 1.54 (6.17-8.36)	2.42± 0.132 (2.33-2.52)
Cantaloupe (n=1)	4.7	5.622
Peaches (n=2)	3.63± 0.57 (3.22-4.04)	16.63 ±3.91 (13.86-19.4)
Clementine (n=1)	3.06	1.25
Pears (n=1)	3.04	1.54
Watermelon (n=1)	0.41	1.286
Vegetables		
Tomato (n=2)	4.08 ±1.38 (3.10-5.06)	3.55± 2.05 (2.11-5.01)
Cucumber (n=1)	0.17	0.157

1. Averages of triplicate determinations of 3 different brands or varieties; Mean ± SD; values in brackets are ranges
2. Prepared by adding 200 mL hot water to 5 g instant coffee granules
3. Prepared by adding 90 mL hot water to 5g ground coffee and boiling for 2 minutes
4. Prepared by adding 200 mL hot water to 2g coffee and steeping for 4 minutes

[a] FW= Fresh weight

n= number of brands or varieties analyzed

Although values obtained by TEAC and FRAP assays differed slightly for a few food items, the results from both assays were highly correlated (r = 0.926, $p < 0.01$) (appendix VIII). This suggests that the compounds capable of reducing oxidants are also responsible for scavenging free radicals in the tested food products.

1.Beverages

The highest antioxidant capacity measured by both assays in the beverages group was exhibited by Instant coffee followed by Turkish coffee, black tea and pineapple canned juice (Table 5) and (Figure 11).

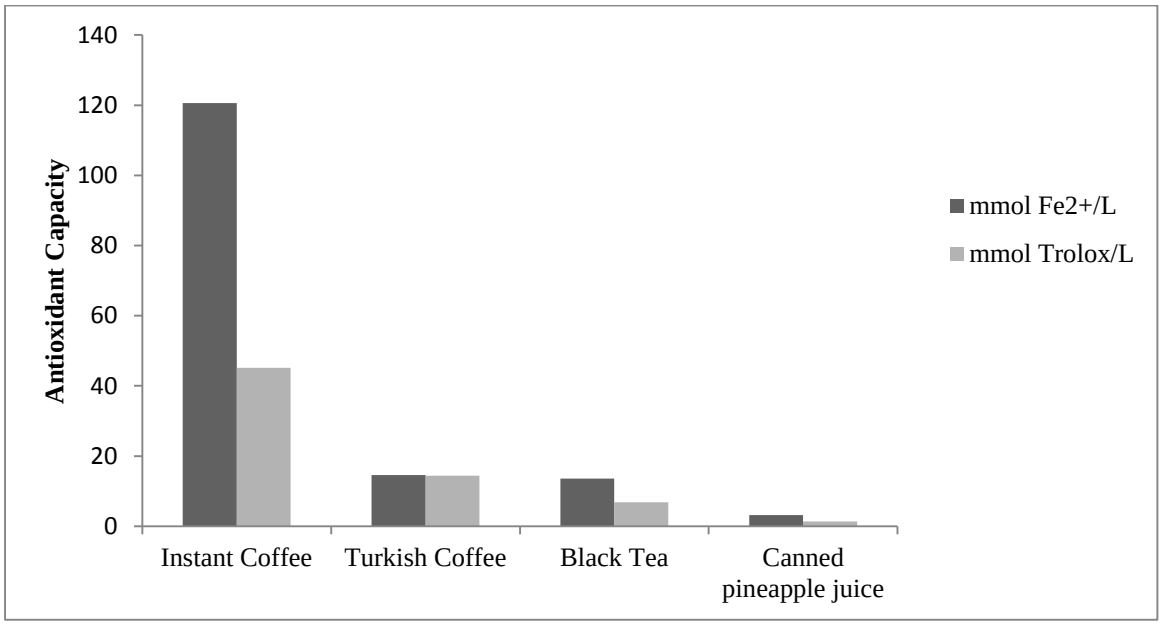


Figure.11 Total antioxidant capacity of the most consumed beverages in Lebanon

a.Instant coffee

- Ferric Reducing Antioxidant Power (FRAP)

The FRAP values of the analyzed brands of instant coffee were in agreement with those reported by other workers (92.7 mmol Fe^{2+}/L, Gorjanović *et al.*, 2017; 108.56 mmol Fe^{2+}/L, Pellegrini *et al.*, 2003). However, it was much higher than that of soluble coffee at 14.17 mmol Fe^{2+}/L (Zujko & Witkowska, 2014).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The mean TEAC values of the analyzed instant coffee brands (45.1 mmol Trolox/L) was similar to those reported for different brands of instant coffee (46.2 mmol Trolox/L, Gorjanović *et al.*, 2017) and soluble coffee consumed in Italy (32.48 mmol Trolox/L, Pellegrini *et al.*, 2003).

b.Turkish coffee

- Ferric Reducing Antioxidant Power (FRAP)

The mean AC value, as measured by FRAP, of the surveyed Turkish coffee brands, was lower than that reported for 13 brand of Turkish coffee (20.75-44.51 mmol Fe^{2+}/L, Gorjanović *et al.*, 2017) and Brazilian ground coffee (62.17 mmol Fe^{2+}/L, Torres& Farah, 2017). However, the mean value was higher than that reported for ground coffee from Poland (5.62 mmol $Fe^{2+/}$L, Zujko & Witkowska, 2014).

Trolox Equivalent Antioxidant Capacity (TEAC)

The TEAC values of the surveyed Turkish coffee brands were within the bracket reported for 13 brands of Turkish coffee (8.56 -22.15 mmol Trolox/L, Gorjanović *et al.*, 2017) and similar to that reported for Brazilian ground coffee (17.86 mmol Trolox/L, Torres& Farah, 2017).

c.Black Tea

- Ferric Reducing Antioxidant Power (FRAP)

The mean total antioxidant capacity (TAC) of different black tea brands measured using FRAP procedure was similar to that of black tea consumed in Italy (10.09 mmol Fe^{2+}/L, Pellegrini *et al*., 2003), higher than that consumed in Poland (7.8 mmol Fe^{2+}/L, Zujko & Witkowska, 2014) and greater than the average value of 5 different brands of black tea including and excluding tea bags (7.22 mmol/L and 8.182 mmol Fe^{2+}/L, respectively; Ryan & Petit, 2010). The different TEAC values of tea with and without the bag indicate an interaction between the bag materials and tea leaves

which affect the antioxidant properties of the product. Other factors which might contribute to the observed differences in antioxidant capacity levels include infusion time, stirring duration and intensity, leaf size, and tea bag porosity (Ryan & Petit, 2010).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The mean TEAC value was similar to that of fermented black tea (5.89 mmol Trolox/L, Bartoszek *et al*., 2018) and higher than that of black teas from Italy (3.60 mmol Trolox/L, Pellegrini *et al.*, 2003) and Poland (0.5-3 mmol Trolox/L; Bancirova, 2010). It is noteworthy that tea catechins are the most powerful antioxidants among the known plant phenols with epigallocatechin-3-gallate (EGCG) reportedly being 20 times more active than vitamin C and 30 times more than vitamin E (Bartoszek *et al.*, 2018).

d.Canned Pineapple Juice

- Antioxidant capacity by FRAP and TEAC

The FRAP and TEAC values of the analyzed commercial pineapple juices in the Lebanese market were similar to those reported for commercial brands in Italy (5.16 mmol Fe^{2+} /L and 1.5 mmol Trolox/L respectively, Pellegrini *et al*., 2003). Furthermore, the TAC measured by the TEAC assay for 13 different brands from the Spanish market ranged between 0.6-15 mmol Trolox/L with a mean value of 3.36 mmol Trolox/L similar to Lebanese processed pineapple juices (Pastoriza *et al.*, 2017).

Solid-phase extraction (Oasis HLB cartridge) was carried out to eliminate the water-soluble reducing interferences. However, results of TAC with and without SPE were the same which indicate that these substances, that interact with Folin-Ciocalteu reagent resulting in overestimated phenol content, doesn't have the same effect on antioxidant capacity.

Fruits and vegetables

Olives exhibited the highest AC values, as determined by the FRAP and TEAC assays, amongst the fruits and vegetables analyzed in the present work (Table 5, Figure 12).

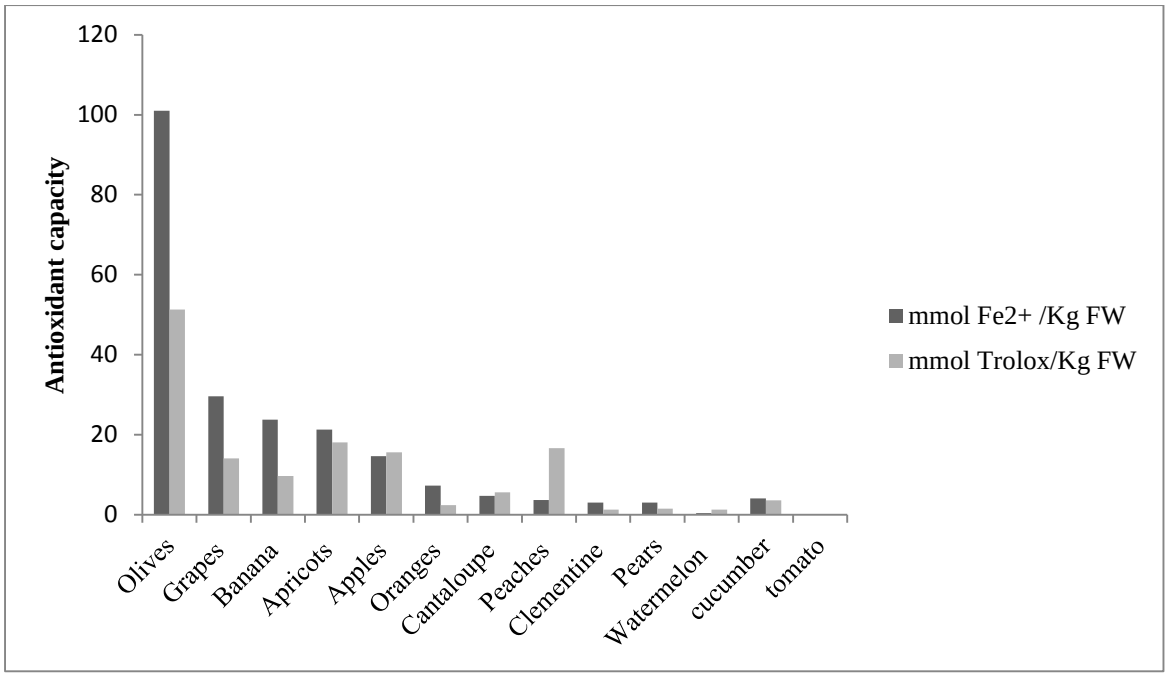


Figure.12 Total Antioxidant Capacity of most consumed Fruits and Vegetables in Lebanon

Results are on fresh weight basis from triplicate analyses of 3 food samples

2.Fruits

a. Olives

Olives had the highest phenol content and antioxidant capacity within the fruits group. Black olives had greater AC than green olives with the "Ayroni" variety having the highest value determined by both assays (Table 6).

Table.6 Total antioxidant capacities for the most consumed table olives

Olives Variety	**mmol Fe^{2+} / Kg FW[a]**	**mmol Trolox/ Kg FW[a]**
Black Olives	154.3 ±127.4 (64.2-244.5)	59.7 ±14.9 (49.1-70.3)
Green Olives	47.7 ±0.76 (47.2-48.2)	42.8 ±19.3 (29.2-56.5)
Mean TAC	101.± 95.9 (47.3-244.5)	51.3± 17.1 (29.1-70.3)

[a.] Fresh weight

- Ferric Reducing Antioxidant Power (FRAP)

The analyzed olive varieties showed greater antioxidant activity compared to those collected from the supermarkets in Guangzhou (China) reportedly having a very low mean AC value (2.7 mmol Fe^{2+} / Kg FW, Fu *et al*., 2011). Further, the assayed black and green olives showed higher values than the black and green olives from the Italian market at 39.9 mmol Fe^{2+} / Kg FW and 24.59 mmol Fe^{2+} / Kg FW, respectively (Pellegrini *et al.*, 2003).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The TEAC value of the surveyed olives were lower than those of Chinese olives (mean of 80.68 mmol Trolox/ Kg FW, Fu *et al.*, 2011) and higher than those of the Turkish variety "Sarıulak" (1.46-2.1 mmol Trolox/ Kg FW, Arslan & Özcan, 2011). Furthermore, the black and green-skinned olives consumed in Italy had lower values at14.73 mmol Trolox/ Kg FW and 10.43 mmol Trolox/ Kg FW, respectively (Pellegrini *et al.*, 2003).

b. Grapes

- Ferric Reducing Antioxidant Power (FRAP)

The FRAP value of the analyzed grape varieties was higher than grapes with red skin (6.70 mmol Fe^{2+} / Kg FW), with green skin (4.95 mmol Fe^{2+} / Kg FW), Jufeng grapes (10.12 mmol Fe^{2+} / Kg FW) and USA grapes (1.73 Fe^{2+} / Kg FW) (Fu *et al*., 2011). Additionally, black and white-skinned grapes from the Italian market exhibited

lower values at 11.09 and 3.25 mmol Fe^{2+} / Kg FW, respectively (Pellegrini *et al.*, 2003).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The local grapes showed higher TEAC value as compared to grapes from other localities. The Chinese market red grapes showed an AC value of 1.27 mmol Trolox/ Kg FW, green grapes 3.95 mmol Trolox/ Kg FW, Jufeng grapes 3.34 mmol Trolox / Kg FW and USA grapes 1.23 mmol Trolox/ Kg FW (Fu *et al.*, 2011). Italian black and white grapes also had lower values at 3.85 and 2.48 mmol Trolox/ Kg FW, respectively (Pellegrini et al., 2003). In contrast, red skin grapes from Spain market showed higher values (20 mmol Trolox/ Kg FW) while white skin grapes had similar levels (13.3 mmol Trolox/ Kg FW) (Garcí *et al.*, 2004).

c. Banana

- Ferric Reducing Antioxidant Power (FRAP)

The FRAP value of bananas varied with the banana variety with the lowest being registered for the Baby banana (3.5 mmol Fe^{2+} / Kg FW) and the highest for Grand Nain (59.03 mmol Fe^{2+} / Kg FW). Bananas available in the Chinese market showed a FRAP value of 5.33 mmol Fe^{2+} / Kg FW for general banana, 3.48 mmol Fe^{2+} / Kg FW for cooking banana and 2.81 mmol Fe^{2+} / Kg FW for royal banana (Fu *et al.*, 2011), while bananas from Italy had 2.28 mmol Fe^{2+} / Kg FW (Pellegrini *et al.*, 2003) and from Brazil 0.7 mmol Fe^{2+} / Kg FW (Torres & Farah, 2017).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The mean AC measured by TEAC was higher than reported by other workers and differed between the analyzed varieties. Baby banana had the lowest TEAC value (2.27 mmol Trolox/ Kg FW) while Grand Nain had the highest (19.98 mmol Trolox/ Kg FW).The antioxidant power of table bananas and cooking bananas were 3.44 and 1.74 mmol Trolox/ Kg FW, respectively, while the royal type showed 1.95 mmol Trolox/ Kg FW (Fu *et al.*, 2011). Moreover, TEAC of bananas from Italy have been reported at 0.64

mmol Trolox/ Kg FW (Pellegrini *et al.*, 2003) and from Brazil at 0.24 mmol Trolox/ Kg FW (Torres & Farah, 2017).

d. Apricot

- Ferric Reducing Antioxidant Power (FRAP)

The AC levels determined by the FRAP assay showed large differences between "Um Hussein" and "Ajami" varieties. "Um Hussein" apricot showed a very high antioxidant activity value that exceeded reported values for apricots from other countries. The FRAP value of the "Ajami" type was similar to apricots from Italy (4.02 mmol Fe2+ / Kg FW, Pellegrini *et al.*, 2003) and higher than those reported for 7 apricot varieties harvested at 3ripening stages (0.045-0.357 mmol Fe^{2+} / Kg FW, Iordanescu *et al.*, 2018).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The two varieties showed higher TEAC values than those reported in literature with "Um Hussein" apricots having double AC value compared to "Ajami" variety. Apricot fruits of nine cultivars, 7 of Italian germplasm and 2 foreign cultivars showed lower values ranging between 1.36 and 4.55 mmol Trolox/ Kg FW (Leccese *et al.*, 2007). Other apricot varieties available in the Italian market had mean AC of 1.44 mmol Trolox/ Kg FW and ranged between 2.69 and 9.01 mmol Trolox/ Kg FW (Pellegrini *et al.*, 2003; Susanna & Annamaria, 2016).

e. Apples

- Ferric Reducing Antioxidant Power (FRAP)

Green (Granny Smith) and yellow-skinned (Golden Delicious) apples showed similar FRAP value which was double that of apples with red peel (Red Delicious) (appendix VI). Similar results were reported for green apples from the Chinese market (9.34 mmol Fe^{2+} / Kg FW, Fu *et al.*, 2011) and higher values were exhibited by those grown in Chile (29.34 mmol Fe^{2+} / Kg FW, Henriquez *et al.*, 2010). Further, yellow apples from Italy showed lower antioxidant activity (3.23 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003) as compared to the yellow apples grown in Lebanon. The local red apples had greater antioxidant power than the red delicious and rose red apples from

China at 5.9 and 9.25 mmol Fe^{2+} / Kg FW, respectively (Fu *et al.*, 2011) and Italy (3.84 25 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003). However, similar results were reported for apples with red skin with AC values ranging between 19.96 and 33.43 mmol Fe^{2+} / Kg FW (Henriquez *et al.*, 2010).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The red apples (Red Delicious) had a TEAC value double that of green (Granny smith) and yellow (Golden Delicious) apples which showed similar antioxidant activity. The TEAC values of the analyzed green and yellow skinned apples were higher than those of their counterparts from China (4.98 mmol Trolox/ Kg FW, Fu *et al.*, 2011) and Italy (1.31 mmol Trolox/ Kg FW, Pellegrini *et al.*, 2003). In addition, the local red apples had higher TEAC than red apples from China & Italy (Fu *et al.*, 2011; Pellegrini *et al.*, 2003).

f. Orange and Clementine

- Ferric Reducing Antioxidant Power (FRAP)

The average FRAP value of local oranges was similar to navel oranges from China (6.02 mmol Fe^{2+} / Kg FW) and USA (9.57 mmol Fe^{2+} / Kg FW (Fu *et al.*, 2011), slightly lower than that from Poland (11.79 mmol Fe^{2+} / Kg FW, Zujko *et al.*, 2012) and lower than that from Italy (20 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003).

The FRAP value of clementine at 3.06 mmol Fe^{2+} / Kg FW was lower than that from Italy (8.88 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003).

- Trolox Equivalent Antioxidant Capacity (TEAC)

Orange fruit showed a mean TEAC that was similar to Navel oranges from China (2.78 mmol Trolox/ Kg FW) and USA (3.72 mmol Trolox/ Kg FW) (Fu *et al.*, 2011), higher than that of Brazilian orange (0.81 mmol Trolox/ Kg FW, Torres & Farah, 2017) and lower than that of Italian types (8.74 mmol Trolox/ Kg FW, Pellegrini *et al.*, 2003).

Clementine had lower AC (1.25 mmol Trolox/ Kg FW) than that from Italy (3.10 mmol Trolox/ Kg FW) (Pellegrini *et al.*, 2003).

g. Cantaloupe

- Antioxidant capacity by FRAP and TEAC

The AC of cantaloupe measured by the FRAP assay (4.7 mmol Fe^{2+} / Kg FW) was similar to values reported by other workers 4.51 mmol Fe^{2+} / Kg FW (Fu *et al.,* 2011) and 5.7 mmol Fe^{2+} / Kg FW (Pellegrini *et al.,* 2003). However, the AC measured using the TEAC assay showed a slightly higher value at 5.62 mmol Trolox/ Kg FW than cantaloupe from the Chinese and Italian markets at 2.56 and 1.20 mmol Trolox/ Kg FW, respectively (Fu *et al.,* 2011; Pellegrini *et al.,* 2003).

h. Peach

- Ferric Reducing Antioxidant Power (FRAP)

Peaches including nectarine showed a FRAP value that is within the bracket (2.5-12.5 mmol Fe^{2+} / Kg FW) reported for eleven peach cultivars (Tavarini *et al.,* 2008) and were similar to peaches consumed in China (2.09 mmol Fe^{2+} / Kg FW, Fu *et al.,* 2011). However, the values were lower than those of yellow peach (6.57 mmol Fe^{2+} / Kg FW, Pellegrini *et al.,* 2003) and the Polish variety (6.9 mmol Fe^{2+} / Kg FW, Zujko *et al.,* 2012).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The mean total antioxidant capacity measured by the TEAC assay was higher than that of yellow peach (1.67 mmol Trolox/ Kg FW, Pellegrini *et al.,* 2003) and peach in China (2.38 mmol Trolox/ Kg FW, Fu *et al.,* 2011).

i. Pear

- Antioxidant capacity by FRAP and TEAC

The FRAP of the local pear variety was similar to that from Poland (3.12 mmol Fe^{2+} / Kg FW, Zujko *et al.,* 2012) and Pear (var. fragrant) in the Chinese market (2.14 mmol Fe^{2+} / Kg FW, Fu *et al.,* 2011). Moreover, it was slightly lower than that reported for the

variety grown in Italy (5 mmol Fe^{2+} / Kg FW, Pellegrini *et al*., 2003). Further, the antioxidant activity measured by the TEAC assay was similar to that of pear (var. fragrant) (1.51 mmol Trolox/ Kg FW, Zujko *et al.*, 2012) and lower than the Italian pear (2.19 mmol Trolox/ Kg FW, Pellegrini *et al*., 2003).

j. Watermelon

- Antioxidant capacity by FRAP and TEAC

The mean AC of watermelon determined by the FRAP method was 0.41 mmol Fe^{2+} / Kg FW similar to that from Brazil (0.45 mmol Fe^{2+} / Kg FW, Torres & Farah, 2017), slightly lower than that from Italy (1.13 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003) and higher than that in China (red pulp) (4.02 mmol Fe^{2+} / Kg FW, Fu *et al.*, 2011). However, the TEAC value (Put value) was higher than the watermelon available in Brazil (0.34 mmol Trolox/ Kg FW, Torres & Farah, 2017) and Italy (0.69 mmol Trolox/ Kg FW, Pellegrini *et al.*, 2003). Further, the watermelon with red pulp from China has been reported to have almost double the TEAC value (2.64 mmol Trolox/ Kg FW, Fu *et al.*, 2011) of the local variety.

3.Vegetables

The polyphenol content and AC, as measured by the FRAP and TEAC assays, were higher for tomatoes than cucumbers in the vegetables group (Figure 12, Table 5).

a. Cucumber

- Ferric Reducing Antioxidant Power (FRAP)

The AC of cucumber as measured by FRAP was 0.17 mmol Fe^{2+} / Kg fresh weight (10.43 mmol Fe^{2+} / Kg dry weight) and was slightly lower than that reported for the same commodity from Brazil (16.3 mmol Fe^{2+} / Kg DW, Tiveron *et al.*, 2012) and Italy (0.71 mmol Fe^{2+} / Kg FW, Pellegrini *et al.*, 2003).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The TEAC values of cucumber were similar to those reported in the literature. The Lebanese cucumber exhibited a TEAC of 0.157 mmol Trolox/ Kg FW (9.87 mmol Trolox/ Kg DW) similar to that for cucumber from Brazil (10.1 mmol Trolox/ Kg DW, Tiveron *et al.,* 2012) and lower than that reported for cucumbers from the Czech Republic (2 mmol Trolox/ Kg FW, Stratil *et al.,* 2006). It is noteworthy that the AC of cucumbers grown in Burkina Faso was not detected by the TEAC assay (Bayili, *et al.,* 2011).

b. Tomato

- Ferric Reducing Antioxidant Power (FRAP)

The mean FRAP value of the analyzed tomato varieties was similar to that reported for tomatoes in Italy consumed either as ingredients in salads (5.12 mmol Fe^{2+} / Kg FW) or as sauce (6.15 mmol Fe^{2+} / Kg FW) and those consumed in the Czech Republic FRAP of 3.5 mmol Fe^{2+} / Kg FW (Pellegrini *et al.,* 2003; Stratil *et al.*, 2006). Lower values were reported for Brazilian Tomatoes (1.17 mmol Fe^{2+} / Kg FW, Torres & Farah, 2017).

- Trolox Equivalent Antioxidant Capacity (TEAC)

The mean AC of tomatoes as measured by the TEAC assay was higher than the values reported for tomatoes from other localities. In Italy, table tomatoes had a TEAC value of 1.65 mmol Trolox/ Kg FW and those processed to sauce a mean TEAC of 1.47 mmol Trolox/ Kg FW (Pellegrini *et al.*, 2003); a TEAC of 1.2 mmol Trolox/ Kg FW, has been reported for tomatoes consumed in Burkina Faso (Bayili, *et al.,* 2011) and 0.37 mmol Trolox/ Kg FW for the commodity from Brazil (Torres & Farah, 2017). Moreover, the AC of several tomato cultivars grown in southwestern Romania ranged from 0.81 mmol Trolox/ Kg FW to 1.74 mmol Trolox/ Kg FW and in Turkey between 0.48 and 1.18 mmol Trolox/ Kg FW (Nour *et al.*, 2013; Erge & Karadeniz, 2011)

C. Non-extractable Proanthocyanidins (NEPA)

Proanthocyanidins (PA) or condensed tannins, a major group of polyphenols, are widely distributed in plant based food. They are oligomers and polymers of flavan-3-ol and flavan- 3, 4-diols. They are divided into extractable PA (EPA) commonly extracted by aqueous solutions of methanol, ethanol or acetone, and non-extractable PA (NEPA) which remain insoluble. NEPA includes PA bounded to the cell wall and associated with proteins that cannot be easily disrupted (Pérez-Jiménez *et al.,* 2009).

The non-extractable proanthocyanidins content in fruits and vegetables is presented in (Table 7). NEPA content in apricot, clementine and olive fruits was not analyzed and studied before. However melon, orange and watermelon showed very low values that were undetected by Pérez-Jiménez unlike ours that showed noticeable results as shown in (Table 7). Similarly, NEPA content in both tomato and cucumber was reported as undetected dissimilar to our results shown in (Table 7) (Pérez-Jiménez & Saura-Calixto, 2015).

Table.7 Non-extractable proanthocyanidins content of the most consumed fruits and vegetables in Lebanon

	Non-extractable Proanthocyanidins	
Fruits	**mg PA/100 g FW[a]**	**mg PA/100 g DW[b]**
Apples (n=3)	58.4 ±30.85 (36.9-93.7)	323.88 ±74.89 (246.2-395.6)
Apricot (n=2)	26.8 ±1.92 (25.4-28.16)	171.92 ±47.58 (138.28-205.5)
Banana (n=3)	14.2 ±5.12 (11.1-20.1)	84.85 ±9.89 (77.2-96)
Cantaloupe	7.76	72.55
Clementine	15.48	138.18
Grapes (n=3)	26.7± 7.46 (21.2-35.2)	188.65 ±70.41 (118.2-259)
Olives (n=4)	62.9± 36.1 (24.4-111.6)	131.44 ± 82.48 (65.87-252)
Orange (n=2)	10.6 ±5.04 (7.09-14.23)	80.68 ±41.91 (51-110.3)

Peaches (n=2)	33.4 ±24.7 (15.9-51)	247.25 ±188.6 (113.8-380.6)
Pears	13.9	92.64
Watermelon	41.95	441.57
Vegetables		
Cucumber	0.59	37.04
Tomato (n=2)	6.42± 3.82 (3.72-9.13)	74.23 ±38.4 (47.06-101.4)

n= number of varieties analyzed

[a] Expressed on Fresh weight

[b] Expressed on Dry weight

1. Fruits

a. Apple

The mean non-extractable proanthocyanidins content in local apples was 58.4 mg PA/ 100 g FW (323.8 mg PA/ 100g DW) similar to that reported by other workers (55 mg PA/ 100 g FW, Zurita *et al.,* 2012; 37-43 mg PA/100g FW, Pérez-Jiménez *et al.,* 2013) and lower than reported for apples consumed in Spain (76 mg PA/100g DW, Pérez-Jiménez & Saura-Calixto, 2015). The Golden delicious apple showed similar results to that reported for the same variety from Spain (383 mg PA/100g DW, Pérez-Jiménez *et al.,* 2009).

b. Banana

The average NEPA contents in local bananas were lower than those reported for bananas in the literature. The reported NEPA contents of bananas spanned a wide range at 980mg/100g DW (Pérez-Jiménez *et al.*, 2009), 1868.3 mg/100g DW (Pérez-Jiménez & Saura-Calixto, 2015) and 1751 mg/100g FW (Zurita *et al.,* 2012).

c. Grape

The NEPA contents in the analyzed grapes were similar to that in white grape (var. Thompson seedles) (168 mg/100g DW, Pérez-Jiménez *et al.,* 2009) and lower than

that in grapes consumed in Europe (294.1 mg/100g DW, Pérez-Jiménez & Saura-Calixto, 2015).

d. Peach

The mean NEPA content in the analyzed peaches was higher than those in European market (148.5 mg/100g DW, Pérez-Jiménez & Saura-Calixto, 2015) and lower than yellow peach with peel (var. Royal) at 676mg/100g DW and nectarine with peel (var. Royal) at 401mg/100g DW (Pérez-Jiménez *et al.*, 2009).

e. Pear

The analyzed pear fruit had higher NEPA content r than that of pear with peel (Abete Fetel) (70 mg/100g DW, Pérez-Jiménez *et al.*, 2009) and much lower than that reported for the fruits consumed in Europe (415.8 mg/100g DW; Pérez-Jiménez & Saura-Calixto, 2015).

D. Total Dietary Phenol Intake

The total dietary phenol intake from beverages, fruits and vegetables was calculated by multiplying the phenol content values of these commodities, as measured by the Folin- Ciocalteu assay, by daily consumption values from the national survey (Table 8). As noted previously, the selected food items covered at least 85% of the total consumption for each food group.

Table 8. Total dietary Phenol intake from most consumed Beverages, Fruits and vegetables in Lebanon

Food Item	**Per capita intake mL or g/day/person[a]**	**Total phenols (mg GAE/ mL or g)**	**Total phenols intake mg/day/person**
Beverages			
Black Tea	153.94	0.988	152.1
Turkish Coffee	32.51	3.698	120.2
Instant Coffee	26.51	3.75	99.4
Pineapple Juice canned	27.01	0.112	3.0
Fruits			
Apples	31.73	2.268	72.0
Grapes	23.1	2.178	50.3
Banana	15.62	0.8	12.5
Watermelon	12.52	0.113	1.4
Pears	12.19	0.404	4.9
Peaches	9.67	0.414	4.0
Oranges	7.257	1.026	7.4
Olives	6.66	9.579	63.8

Cantaloupe	4.62	0.357	1.6
Apricots	4.4	1.099	4.8
Clementine	3.72	0.725	2.7
Vegetables			
Tomato	29.66	0.413	12.2
Cucumber	24.91	0.0439	1.1
Total			613.6

[a] Chamieh et al., 2015

Beverages were the largest source of dietary phenols (61%) followed by fruits (37%) and vegetables that accounted for only 2.16% (Figure 13). However, the largest single source of TP was Black Tea at 24.8 %, followed by Turkish coffee, Instant coffee, apples, olives and grapes at 19.6, 16.2, 11.7, 10.4 and 8.2% respectively (Figure 14).

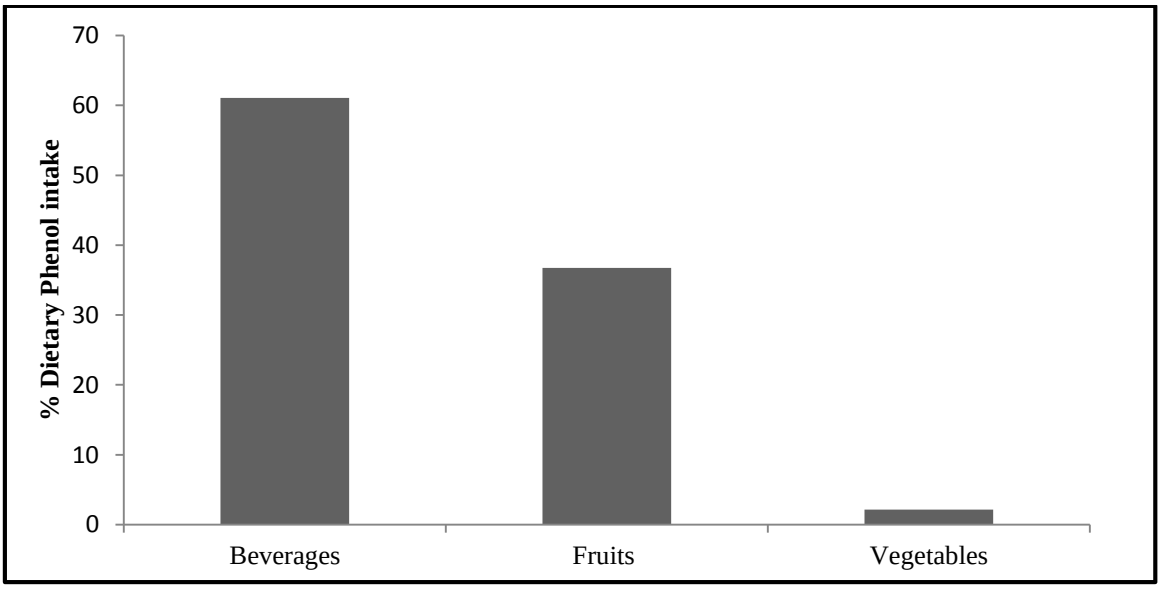


Figure 13. The percent of Dietary phenol Intake from Beverages, Fruits and Vegetables

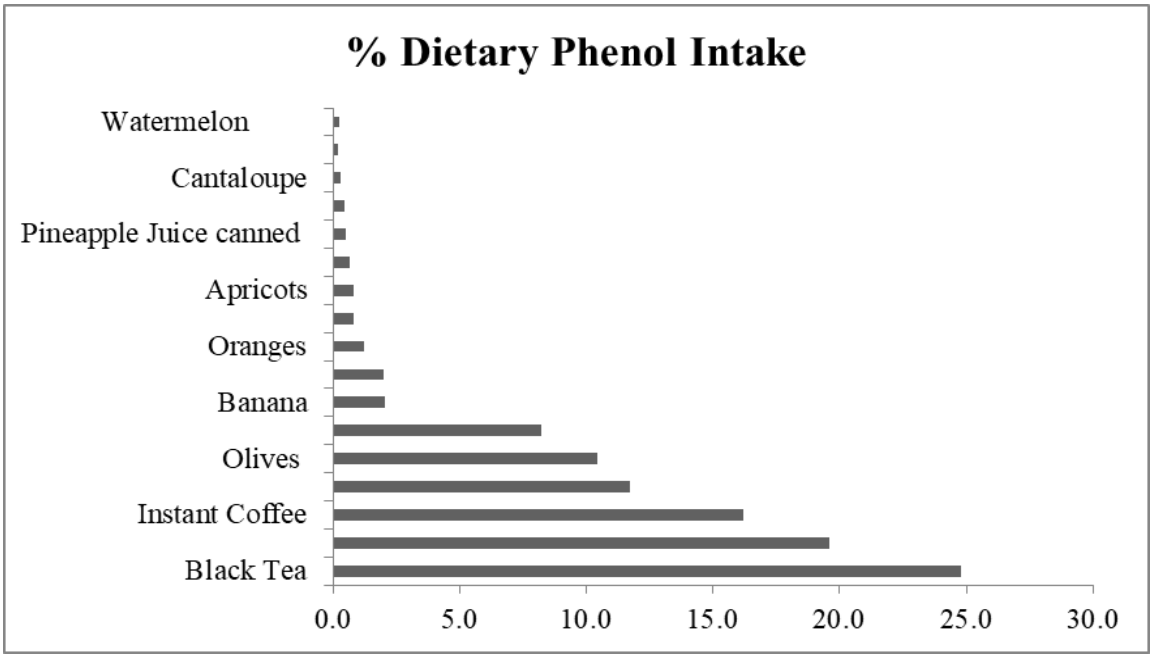


Figure 14. Percentage of Dietary phenol intake in most consumed food Items

The mean consumption of fresh fruits and raw vegetables combined was estimated at 186 g day^{-1} which is lower than the FAO/WHO recommended intake of 400 g of fruits and vegetables per day (Jones & Charlton, 2014). Further, the Lebanese daily intake of phenols from beverages, fruits and vegetables was estimated at 613.4 mg GAE with beverages accounting for 374.7 mg GAE, fruits at 225.4 mg GAE and vegetables at 13.3 mg GAE.

The daily phenol intake from beverages in Lebanon was lower than that from beverages consumed as part of the Polish diet (484 mg GAE, Zujko & Witkowska, 2014) and the Spanish diet (614 mg GAE, Saura-Calixto & Goñi, 2006) presumably due to higher consumption of wine in Poland and Spain. Further, Japanese middle-aged women had higher dietary polyphenol intake from beverages at 664.4 mg GAE due to higher coffee and tea consumption (Fukushima *et al.*, 2014). The above mentioned studies differed in methods used for determining food intakes, as for that done in Japan participants tended to record their daily dietary intake over a period of 7 consecutive days while in Spain 7 non-consecutive days were included. Being recorded by participants with no supervision can lead to under- or over-estimation of the dietary intake. Furthermore, the study done in Poland utilized a single 24 hour recall for determining the dietary intake.

Moreover, the Lebanese daily phenol intake from both fruits and vegetables accounted for 238.7 mg GAE lower than the intake in France from fruit and vegetables (280 mg GAE, Brat *et al.,* 2006) and Spain at 295 mg GAE with fruits and vegetables accounting for 178 and 117 mg GAE respectively (Saura-Calixto & Goñi, 2006). Moreover, This value was also lower than the American daily intake estimated at 450mg GAE with 320.4 mg GAE for fruits and 129.4 mg GAE for vegetables (Chun *et al.,* 2005) and Polish intake at 451.5 mg GAE having lower PP intake from fruits (196.5 mg GAE) and much higher from vegetables at 266 mg GAE (Zujko & Witkowska, 2014) as compared to these in Lebanon.

E. Antioxidant capacity of beverages, fruits and vegetables

The antioxidant capacity of the beverages, fruits and vegetables commonly consumed in Lebanon was evaluated by two *in vitro* assays, viz. FRAP and TEAC. These two assays were chosen to measure different mechanisms of antioxidant action and to allow comparison with the literature data. FRAP assay is based on single electron transfer (SET) while TEAC assay is based on both hydrogen atom transfer (HAT) and single electron transfer (SET). Moreover, the TEAC assay is sensitive to lipophilic and hydrophilic antioxidant compounds while FRAP is more sensitive to hydrophilic compounds. Some antioxidants can be classified as hydrophilic (e.g. ascorbic acid) or lipophilic (e.g. carotenoids and vitamin E) compounds. However, it is difficult to separate polyphenols into hydrophilic and lipophilic because they are complex group substances with different molecular mass and can be either free or bound to protein or dietary fiber (Pulido *et al.,* 2003).

The evaluated food products were classified according to the AC as very high, high, medium or low, considering the percentiles' distribution of AC values. Values within the first quartile (25^{th}) were classified as low contributors while those within second quartile (50^{th}) and third quartile (75^{th}) were considered to be of medium contribution. AC values above the third quartile were classified as high and very high when values are above the 90^{th} percentile (Table 9).

Table.9 Antioxidant capacity (AC) of the most frequently consumed beverages, fruits and vegetables in Lebanon

Classification	**Food product**	**TEAC mmol Trolox/L or Kg**				**FRAP mmol Fe^{2+}/L or Kg**			
		Mean	**± SD**	**%**	**Rank**	**Mean**	**± SD**	**%**	**Rank**
Very High	Instant Coffee	45.1	8.04	22.2	**1**	120.6	83.2	33.2	**1**
	Olive	45.04	12.4	22.2	**2**	95.6	90.3	26.3	**2**
	Apricot	18.1	7.63	8.93	**3**	21.28	25.7	5.86	**5**
	Peach	16.63	3.91	8.21	**4**	3.63	0.57	1.00	**12**
High	Apples	15.6	8.7	7.70	**5**	14.6	6.35	4.02	**7**
	Turkish Coffee	14.35	0.42	7.08	**6**	14.6	0.35	4.02	**6**
	Grape	14.1	4	6.96	**7**	29.6	2.96	8.15	**3**
	Banana	9.69	9.19	4.78	**8**	23.77	30.6	6.54	**4**
Medium	Black Tea	6.82	1.1	3.3	**9**	13.6	6.3	3.7	**8**

				7				4	
	Cantaloupe	5.622	…	2.77	**10**	4.7	…	1.29	**10**
	Tomato	3.55	2.05	1.75	**11**	4.08	1.38	1.12	**11**
	Orange	2.42	0.132	1.19	**12**	7.266	1.54	2.00	**9**
	Pear	1.54	…	0.76	**13**	3.04	…	0.84	**15**
Low	Pineapple Canned Juice	1.39	0.7	0.69	**14**	3.19	2.11	0.88	**13**
	Watermelon	1.286	…	0.63	**15**	0.41	…	0.11	**16**
	Clementine	1.25	…	0.62	**16**	3.06	…	0.84	**14**
	Cucumber	0.157	…	0.08	**17**	0.17	…	0.05	**17**

Instant coffee showed very high AC when measured by both assays. Coffee is known to be a rich source of compounds with powerful antioxidant activity due to its high contents of polyphenols and melanoidins (Contreras-Calderón *et al.,* 2016). The high AC and potential beneficial health effects of coffee consumption have been reported in many epidemiological and clinical *in vitro* studies. Coffee consumption and antioxidant effect have been associated with lower incidence of chronic degenerative diseases such as coronary diseases, type 2 diabetes and some types of cancers including colon and liver cancer (Torres & Farah, 2017). The AC of coffee is affected by many factors including variety and origin of coffee, roasting degree and type and technological parameters used in coffee brew extraction (Contreras-Calderón *et al.,* 2016). Instant coffee had higher AC compared to Turkish coffee because of the formation of melanoidins during roasting in ground coffee. Melanoidins are probably responsible for the low response of scavenging activity in ground coffee that are usually

prepared from dark roasted coffee beans unlike instant coffee that is prepared from light medium roasted beans. Moreover, some compounds of coffee are volatile and might be evaporated during boiling process in Turkish coffee (Al Doghaither *et al.*, 2017).

Olives, apricot and peaches showed very high AC when measured by TEAC assay. On the contrary, apricots and peaches showed a high and medium AC respectively by FRAP assay while olives showed very high AC by both assays. This suggests that both apricots and peaches have higher lipophilic antioxidant (e.g. carotenoids) compounds than hydrophilic ones.

The high AC of grapes is mainly derived from the high anthocyanin and flavonoids content and proanthoycanin especially in the red-skinned grapes (Xia et al., 2010). Oranges showed medium AC value due to their content of vitamin C and flavonols (Torres& Farah, 2017). The high AC of apples and bananas could be attributed to their high contents of phenolic acids and flavanols. Black tea showed medium AC possibly due to the loss in antioxidant capacity during the process of fermentation where the flavanols in green tea leaves undergo oxidative polymerization by polyphenol oxidase (Pellegrini *et al.*, 2003). Unexpectedly, tomato showed a medium AC as measured by both assays. Tomatoes are rich in lycopene which is a carotenoid pigment that can quench various reactive species including singlet oxygen (Torres & Farah, 2017). This indicates that either the AC assays used do not use singlet oxygen in their oxidation protocol or there is incomplete extraction of lycopene from tomato being present in crystalline form in the chromoplast thereby reducing the extraction efficiency by acetone.

F. Estimation of the contribution of beverages, fruits and vegetables to antioxidant intake

The average percent contribution of each food group to Lebanese' dietary AC, based on the AC data obtained by TEAC and FRAP assays associated with the consumption database, is presented in Figure 15.

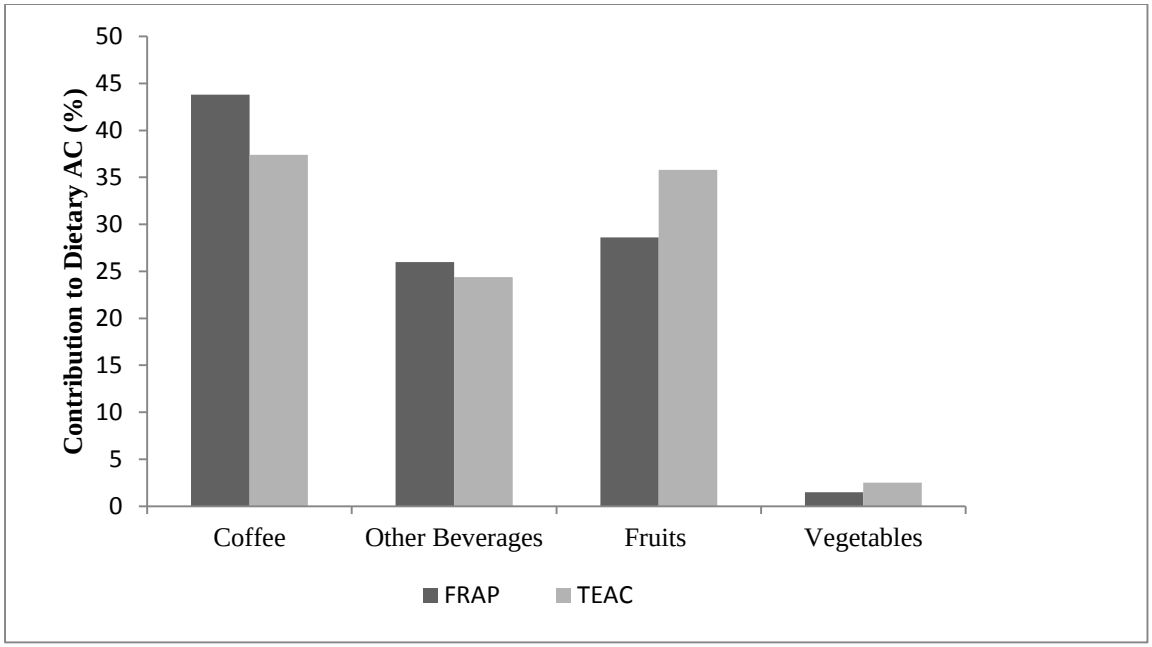


Figure 15. Percent contribution of coffee and the remaining food groups to the dietary antioxidant capacity (AC) of the analyzed foods

No significant difference was observed in dietary AC when TEAC and FRAP assay results were considered (paired T test, $p > 0.05$). Speaking of the country as a whole, although other beverages (black tea and pineapple canned juices), fruits and vegetables also contributed to the dietary AC, their percentage contributions were considerably lower than that of coffee (Figure 15). Coffee had the highest average contribution to the dietary AC (40.6%), followed by fruits (32.2%) and other beverages (black tea and pineapple juice) which contribute 25.2% to antioxidant intake. Vegetables contributed very little (2%) to the antioxidant intake from the surveyed foods.

Instant coffee, black tea, apples, Turkish coffee, grapes and olives are the main contributors to the total dietary AC in Lebanon. The importance of coffee to the dietary

AC in Lebanon was expected, given its high consumption in the country. Lebanon consumed approximately 26,417 tons of coffee in 2016 with Lebanese Coffee (Turkish coffee) accounting for 65% of total consumption of coffee followed by instant coffee at 35% (Mikhael, 2017). Instant coffee exhibited higher DAC compared to Turkish coffee due to its higher AC as measured by the FRAP and TEAC assays. The high contribution of black tea to DAC is due to its high consumption (Chamieh *et al.,* 2015) and high AC (Table 5). Likewise apples, grapes and olives are highly consumed in Lebanon as apples account for 23% and grapes 14% of total fruit production while olives cover around 21% of the total cultivated area (IDAL 2015).

CHAPTER V

CONCLUSION AND RECOMMENDATION

Non-communicable diseases are estimated to account for 91% of all deaths in Lebanon and their burden have been rising over the last century (World Health Organization, 2018).Of the NCDs, cardiovascular diseases account for 47% while cancer for 16% and diabetes for 5% (World Health Organization, 2018). Exploring dietary factors influencing these conditions became important in order to design effective strategies for reducing the burden of these diseases. Several studies have demonstrated the association between oxidative stress and NCDs (Grosso, 2018). Notwithstanding the important role of antioxidants in curtailing the development of many pathological conditions, few studies quantified the dietary phenol intake. This study is the first to address the phenol contents and antioxidant intakes of phenols in Lebanon and the MENA region and underscores the need of such studies for increasing consumer's awareness and curbing the scourge of NCDs in this part of the world.

The Lebanese daily intake of phenols from the most consumed beverages, fruits and vegetables was estimated at 613.4 mg GAE. Beverages accounted for 374.7 mg GAE, fruits for 225.4 mg GAE and vegetables for 13.3 mg GAE. The largest single source of TP was black tea, followed by Turkish coffee, instant coffee, apples, olives and grapes.

In the beverages group, instant coffee and Turkish coffee, as prepared in this study, ehibited the highest phenol content at 375 mg/100ml and 369.8 mg/100ml, respectively. While in fruits group, table olives showed the highest phenol content especially black olives at 957.9 mg/100g FW followed by apples at 226.8 mg/100g FW and grapes at 217.8 mg/100g FW. Red- skinned apples and grapes exhibited higher phenol content than the other varieties.

Instant coffee, black tea, apples, Turkish coffee, grapes and olives are the main contributors to the total dietary AC in Lebanon. The dietary antioxidant capacity was calculated by multiplying the AC values of the analyzed commodities by the daily per capita consumption values from the national survey database. The antioxidant capacity (AC) was measured by two assays, viz. the ferric reducing antioxidant power (FRAP)

and the trolox equivalent antioxidant capacity (TEAC). The AC value of the surveyed food items ranged between 0.17 and 120.6 mmol Fe^{2+} /L or Kg FW as assessed by the FRAP assay and between 0.157 and 45.1 mmol Trolox/L or Kg FW by TEAC. Higher FRAP value suggests that hydrophilic antioxidant compounds are more prevalent than the lipophilic compounds. A high and positive correlation was obtained between the total phenol content of the analyzed food items and antioxidant capacity measured by FRAP assay ($r = 0.779$, $p < 0.01$) as well as the TEAC assay ($r = 0.867$, $p < 0.01$). This indicates that phenolic compounds are the main modulators of the reducing ability and free radical- scavenging capacity of the analyzed commodities.

The commodities with the highest AC as measured by the TEAC assay were instant coffee (120.6 mmol Trolox /L), olives (101 mmol Trolox / Kg FW), apricot (18.1 mmol Trolox / Kg FW), peach (16.63 mmol Trolox / Kg FW) and apples (15.6 mmol Trolox / Kg FW). Likewise, instant coffee, olives and apricot had the highest AC value as determined by the TEAC assay at 45.1, 51.3 and 25.7 mmol Fe^{2+}/L (or Kg) FW, respectively. Grapes (29.6 mmol Fe^{2+} /Kg FW) and bananas (23.77 mmol Fe^{2+} / Kg FW) ranked amongst the top 5 commodities in AC as assessed by the FRAP assay. Although values obtained by TEAC and FRAP assays differed slightly for a few food items, the results from both assays were highly correlated ($r = 0.926$, $p < 0.01$). This suggests that the compounds capable of reducing oxidants are also responsible for scavenging free radicals in the tested food products.

The total non-extractable proanthoycanins (NEPA) content of fruits and vegetables was estimated at 319.1 mg PA/100g FW with the highest content being in olives, apples and watermelon at 62.9, 58.4, and 41.9 mg PA/100g FW, respectively. The NEPA content in apricot, clementine and olives was analyzed for the first time in this study.

Findings from this study highlight the importance of increasing the Lebanese vegetable and fruit daily intakes to reach the value recommended by FAO/WHO of 400 g/ day to increase total phenols intakes in Lebanon. The low TP content of the cultivated cucumber variety, warrant the introduction of varieties expressing higher levels of TP.

Moreover higher olive and apple intakes, especially black olives (var. Ayroni) and red skinned apple (var. Red delicious), is recommended in view of the relatively-higher levels of TP and AC. It is also recommended to focus more on fruit and vegetable varieties that are rich in TP and exhibit high AC such as Grand Nain bananas, cherry tomato, Maghdoushi red grapes, and Um Hussein apricots. Also increasing the daily intake of coffee (Turkish and instant) is recommended with the caveat of remaining within the caffeine daily limit. Further, fresh fruit juices are recommended over processed ones because processing of fruit juices is often accompanied by a decrease in their TPC.

Further studies are needed to determine non-extractable polyphenols NEPP content (HPP and NEPA) and identify their AC in most consumed fruits and vegetables. Further, investigation the TPC and AC of the most consumed Lebanese composite dishes would provide useful insights on antioxidant intakes from the Lebanese diet. Moreover, studies that investigates the relationship between antioxidant intake and factors including: age, gender, socioeconomic status and geographic region would further give a better view of the antioxidant status and suggest more robust recommendations.

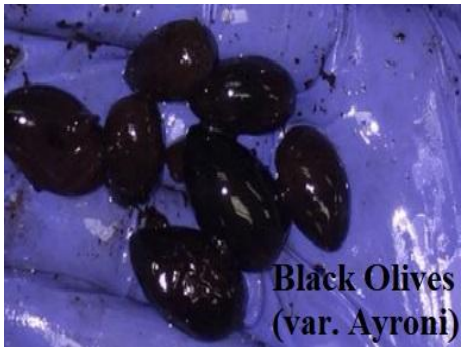
Strength and Limitations

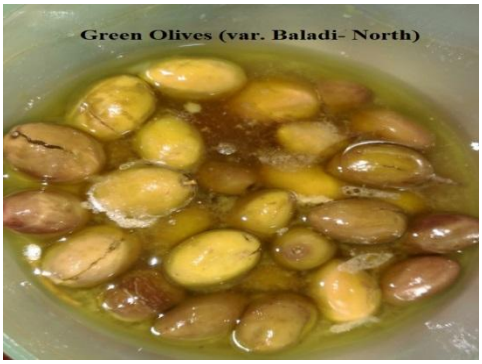
The study is the first in MENA region to determine dietary phenol intake through quantifying phenol content and antioxidant capacity of most consumed fruits, vegetables and beverages in Lebanon. Further, this study quantified the non-extractable proanthocyanidins which are usually neglected in research work in spite of their positive and significant health effects. Moreover, two different in vitro assays were used to quantify antioxidant capacities which are based on the two major routes through which antioxidant molecules exert their action.

The two major limitations of this study were the approach followed in gathering the food consumption data which was the 24 hour recall which reflects short term intake and that the non-extractable hydrolysable polyphenols (HPP) weren't quantified for a fuller comparison between the contents of extractable and non-extractable polyphenols and their potential health effects .

APPENDIX I

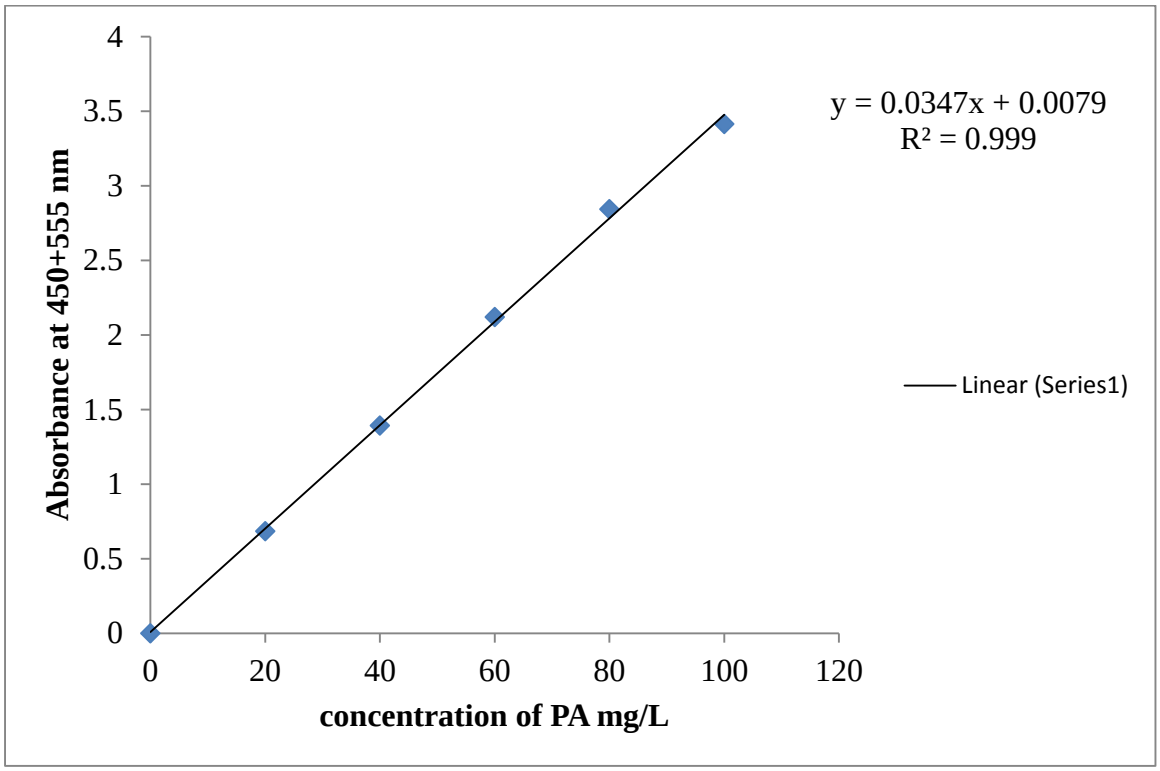
SELECTED TURKISH COFFEE BRANDS AND OLIVE VARIETIES





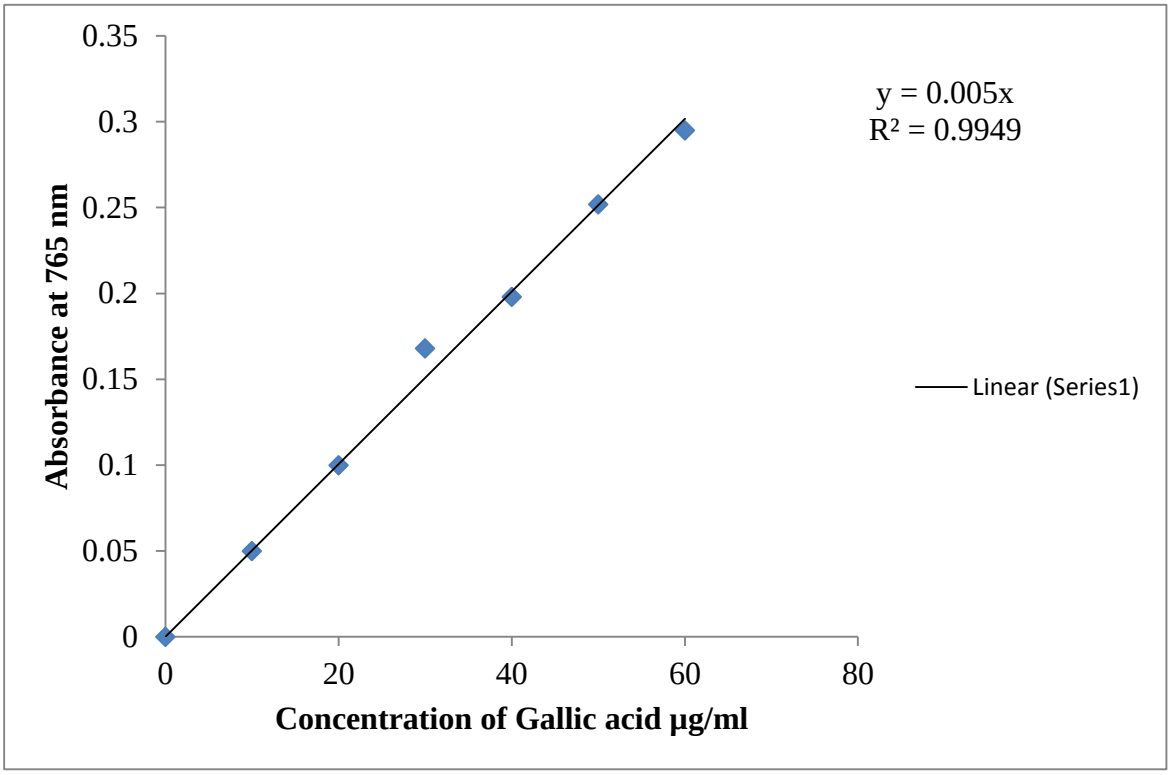
APPENDIX II

STANDARD CURVE FOR NEPA FROM CAROB CONCENTRATE



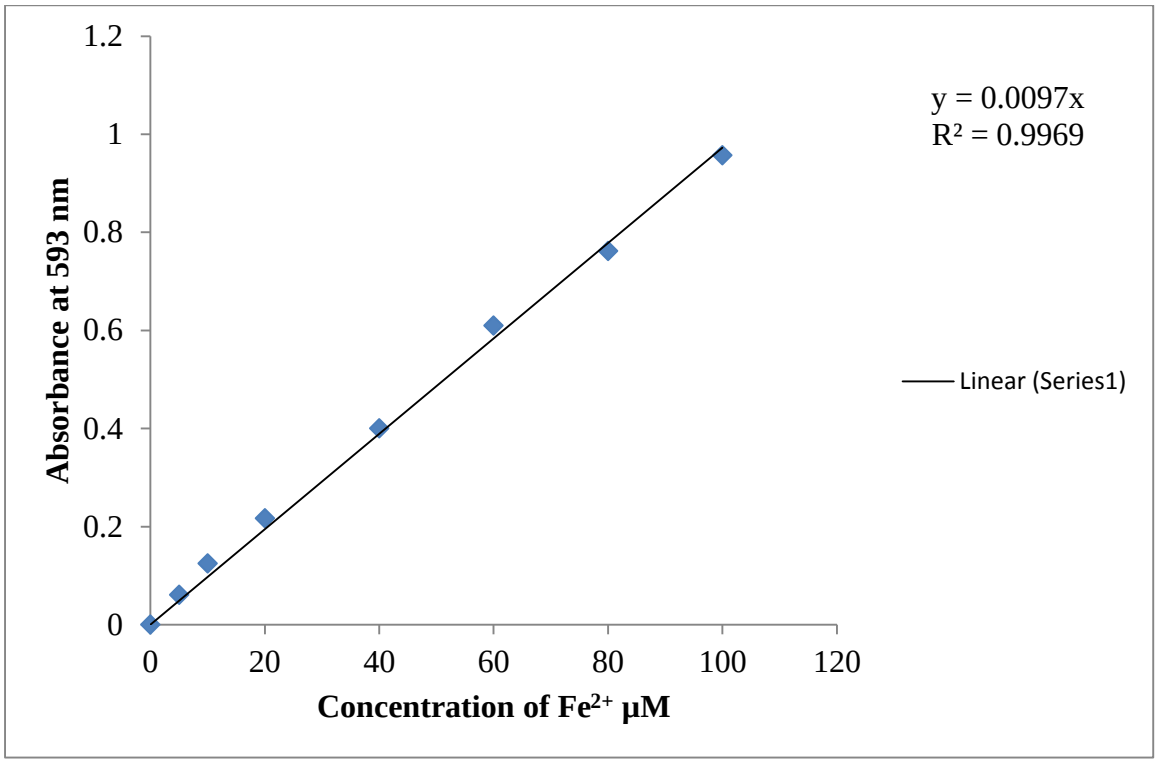
APPENDIX III

GALLIC ACID STANDARD CURVE



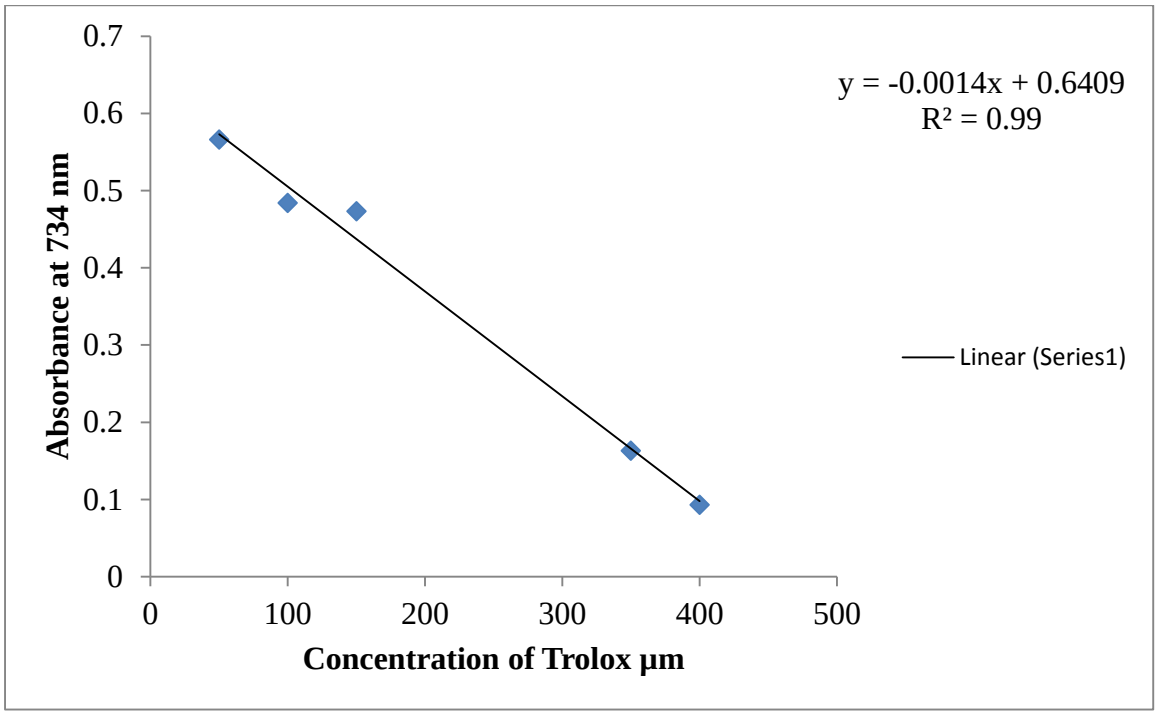
APPENDIX IV

FERROUS SULFATE STANDARD CURVE



APPENDIX V

TROLOX STANDARD CURVE



APPENDIX VI

DETAILED TABLE FOR TPC AND AC CONTENT IN ALL ANALYZED BRANDS AND VARIETIES

	Brand & Variety	TPC mg/100ml or 100g Fresh wt		FRAP mmol/L orKg Fw		TEAC mmol/L or kg Fw	
		Mean	± SD	Mean	± SD	Mean	± SD
BEVERAGES							
Black Tea	Ahmad	94.26	11.57	10.44	0.72	7.236	0.19
2g/200 ml	Horse Head	99.86	28.95	20.96	0.74	5.57	0.34
	Lipton	102.53	15.85	9.44	1.45	7.67	0.09
Coffee	Maatouk	368.66	2.31	14.66	0.92	13.87	0.25
5g/90 ml	Najjar	376	5.40	15.00	1.85	14.67	0.30
	Daniel café	365	5.32	14.30	1.20	14.51	0.27
Instant coffee	Nescafe` Red mug	315	18.80	32.80	0.45	54.36	3.61
5g/200 ml	Nescafe` Gold blend	395	10.89	130.97	1.68	41.4	1.59
	Nescafe matinal	415	8.66	198.26	5.99	39.63	0.77
Pineapple Juice canned	Tropicana						
	Raw Extract RE	15.56	4.82	1.810	0.140	0.89	0.05
10g/10 ml	Washing Extract WE	32.10	5.40				
	Total: WE-RE	16.60					
	Maccaw						
	RE	40.30	2.96	5.63	1.20	2.2	0.06
	WE	51.10	4.45				
	Total: WE-RE	10.80					
	Xtra						
	RE	17.93	1.10	2.139	0.102	1.08	0.045
	WE	24.30	5.30				
	Total: WE-RE	6.37					
VEGETABLES							
Cucumber	Lebanese	4.39	0.38	0.167	0.001	0.157	0.004

Tomato	Green house	27.6	1.72	3.1	0.099	2.107	0.242
	cherry tomato	55.08	3.02	5.06	0.02	5.01	0.21
	FRUITS						
Banana	Baby banana	59.51	9.96	3.52	0.68	2.27	0.007
	Cavendish	32.35	7.01	8.72	2.41	6.83	0.49
	Grand nain	148.2	1.67	59.03	2.59	19.98	0.05
Orange	Common orange (shamouti)	99.48	8.12	8.36	0.743	2.518	0.19
	Navel (abo sora)	105.8	10.3	6.17	0.355	2.33	0.53
Clementine		72.51	6.27	3.06	0.192	1.257	0.185
Pears		40.5	2.16	3.04	0.183	1.54	0.06
Apples	Golden yellow	158.8	4.21	10.78	0.07	9.35	1.14
	Granny smith	156.96	4.86	11.08	0.65	11.95	1.94
	Red Delicious	364.9	2.68	21.94	0.269	25.65	4.5
Grapes	Beitamouri- white	183.87	4.74	29.94	2.74	10.77	0.517
	Helwani -red	186.9	5.74	26.5	1.41	13.21	0.402
	Maghdoushi	282.9	2.02	32.4	6.08	18.6	0.505
olives	Ayroni black	1733.01	150.35	231	7.36	53.1	8.59
	Baladi black	1120.8	37.3	56.1	6.71	41.49	1.01
	Baladi green	449.6	6.29	47.3	3.87	29.1	0.867
	Baladi green north	528.3	7.14	48.25	0.27	56.5	1.03
Watermelon	Crimson-oblong	11.32	0.86	0.411	0.01	1.285	0.085
Peaches	Babcock yellow	38.82	4.01	3.22	0.22	13.86	1.86
	Nectarine	44.05	4.42	4.04	0.56	19.4	0.27
Apricot	Ajami	33.56	4.67	3.08	0.209	12.70	0.26
	Um hussein	186.3	10.84	39.48	7.43	23.50	2.1
Cantaloupe	Ananas	35.69	2.28	4.7	0.25	5.622	0.291

APPENDIX VII

TABLE OF CORRELATION BETWEEN TPC AND AC

Correlation between TPC and FRAP

		TPC	FRAP
TPC	Pearson Correlation	1	.779**
	Sig. (2-tailed)		.000
	N	17	17
FRAP	Pearson Correlation	.779**	1
	Sig. (2-tailed)	.000	
	N	17	17

**. Correlation is significant at the 0.01 level (2-tailed).

Correlation between TPC and TEAC

		TPC	TEAC
TPC	Pearson Correlation	1	.867**
	Sig. (2-tailed)		.000
	N	17	17
TEAC	Pearson Correlation	.867**	1
	Sig. (2-tailed)	.000	
	N	17	17

**. Correlation is significant at the 0.01 level (2-tailed).

APPENDIX VIII

TABLE OF CORRELATION BETWEEN FRAP AND TEAC ASSAYS

Correlations between FRAP and TEAC

		FRAP	TEAC
FRAP	Pearson Correlation	1	.926**
	Sig. (2-tailed)		.000
	N	17	17
TEAC	Pearson Correlation	.926**	1
	Sig. (2-tailed)	.000	
	N	17	17

**. Correlation is significant at the 0.01 level (2-tailed).

APPENDIX IX

MOISTURE CONTENT OF THE ANALYZED FOOD ITEMS

Food item	Varieties	%Moisture content
Cucumber	Baladi	98.4
Banana	Baby	88.4
	Cavendish	75.19
	Grand Nain	85.18
Tomato	Green House	92.1
	Cherry Tomato	91
Orange	Navel	87.1
	Common Orange	86.1
Clementine	Baladi	88.8
Pears	Baladi	85
Apples	Granny Smith	86.5
	Red Delicious	76.3
	Golden	85
Grapes	Helwani -Red	87.3
	Beitamouni- White	86.4
	Maghdoushi	82.14
olives	Baladi Black	40
	Baladi Green	62.9
	Ayroni Black	55.7
	Green North	48.4
Waterm**elon**	Crimson-Oblong	90.5
Peaches	Nectarine	86.6
	Babcock Yellow	86
Apricot	Um Hussein	86.3
	Ajami	81.6
Cantaloupe	Ananas	89.3

APPENDIX X

THE FOOD CONSUMPTION DATA OBTAINED FROM THE NATIONAL SURVEY

Food product	**Intake in g or mL/day/ person**	**Per capita annual intake in kg or L**	**Percentage**
Beverages			
Black Tea	153.94	56.19	55.5
Turkish Coffee	32.51	11.87	11.7
Pineapple Canned Juice	27.01	9.86	9.73
Instant Coffee	26.51	9.68	9.55
Total			**86.5**
Fruits			
Apples	31.73	11.6	20.68
Grapes	23.1	8.4	15.06
Banana	15.62	5.7	10.18
Watermelon	12.52	4.6	8.16
Pears	12.19	4.4	7.95
Oranges/ Clementine	10.99	4.0	7.16
Peaches	9.67	3.5	6.30
Olives	6.66	2.4	4.34
Cantaloupe	4.62	1.7	3.01
Apricots	4.4	1.6	2.87
Total			**85.7**
Vegetables			
Tomato	29.66	10.8	47.1
Cucumber	24.91	9.1	39.6
Total			**86.6**

*Chamieh *et al.*, 2015

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