

ROOFTOP VEGETABLES AND URBAN  
CONTAMINATION: TRACE ELEMENTS AND  
POLYCYCLIC AROMATIC HYDROCARBONS IN CROPS  
FROM HELSINKI ROOFTOPS

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Tiivistelmä – Referat – Abstract <p>The increased rates of population growth and urbanization worldwide raises the question of food security and self-reliance in cities. In view of this situation, in recent years there has been a re-emergence of urban agriculture in its traditional form and in new variations, such as on urban rooftops. A number of rooftop urban farms exist in the world; however, very few studies have been done to establish the quality of crops they produce, specifically concerning the concentrations of contaminants.</p> <p>The main purpose of this study was to investigate levels of contamination in edible plants grown on urban rooftops. I determined concentrations of polycyclic aromatic hydrocarbons (PAH) and trace metals in the biomass of three types of horticultural crops grown in the city of Helsinki, Finland. Lettuce, radish and peas were planted on five rooftops in various areas of Helsinki and control samples were acquired from local food stores and markets. Both groups of crops were analyzed for concentrations of 11 trace elements using the Elan 6000 ICP-MS and 16 PAHs using Shimadzu GC-MS-QP2010 Ultra system with the AOC-20i /AOC-20s autosampler. Additionally, lettuce and pea samples from the roofs were analyzed washed and unwashed to establish levels of particulate contamination on the surface of plants that can be mechanically removed through washing.</p> <p>Results obtained suggest that concentrations of PAHs and trace metals in rooftop vegetables in Helsinki are very low and the differences in their concentrations compared to control (store) samples are insignificant. This demonstrates that the consumption of vegetables produced in uncontaminated soil on urban roofs in Helsinki is safe. All samples showed concentrations well below the safety limits for heavy metals and PAHs established in the European Union (EC, 2006). Finally, there was a difference in concentration of PAHs and trace metals between washed and unwashed samples, however most of the results did not show statistical significance.</p>			
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## **1. Introduction**

### **1.1. General background**

Modern cities are expanding, becoming denser, higher and wider. More people now live in cities than rural areas worldwide, and the majority of future population growth is expected to take place in urban centers (Sharanbir & Parwinder 2012). The rate of global urbanization is estimated to reach 70% by 2050 (UN 2008), and by 2015, the world is expected to have 564 cities with a population of over a million, and 425 of those cities will be in developing countries (Mougeut 2006). The rapid increase in the urban population will place a strain on all areas of city functions and will create a new wave of urban poverty: the UN-HABITAT (2004) estimates that poverty in cities of low-middle income countries will increase from 30-50% by 2020. Since modern cities rely almost exclusively on external resources, including food, the expected rapid urban growth might cause food shortages and put the health and wellbeing of their citizens at risk. Based on population growth rates, estimations show that by 2050, the world will need 1 billion hectares more of arable land to feed itself, which is around the area of Brazil (Despommier 2010).

In light of this rapid urbanization, there is a concern about urban food security worldwide, which was formally discussed at a G8 meeting in Italy in 2009 (Morgan & Sonnino 2010). Increasingly, there is an emphasis on local food production that could slightly ease cities' dependencies on external sources for provisions, alleviate urban poverty and nutrition problems, as well as reduce cities' ecological footprint (Nelson 1996, Mougeut 2006). Producing and consuming food locally could reduce current rate of world food loss, which is currently around 1.3 billion tons per year (Gustavsson et al. 2011). Significant amounts of food are wasted at the consumer level, however, most of the loss is due to spoilage that happens during storing, packaging, transportation and distribution - all of which could be minimized if food were to be consumed in the

area where it was grown (Gustavsson et al. 2011). The less food is wasted, the less of it would need to be produced, reducing the overall environmental impact. This is demonstrated in Doron's (2005) study, where he calculates that if the United Kingdom produced and consumed food locally, it could reduce the level of its carbon dioxide emissions by 22%, which is twice the amount agreed upon under the Kyoto Protocol (Sharanbir & Parwinder 2012).

Urban agriculture is considered an important solution to food security in the increasingly urbanized world. Rooftops are a new addition to possible places to grow vegetables and fruit, house honey bees, and even have small animals like chickens, rabbits and fish (Doron 2005, Mougeut 2006). Urban land is increasingly limited and expensive, and urban rooftops could potentially provide space for agriculture to alleviate space related issues (Whittinghill & Rowe 2011). This type of food production is gaining popularity throughout the world and is becoming a trend in cities of high population density and land prices such as New York, Toronto, Berlin, Tokyo and Hong Kong, to name a few (Doron 2005).

In the Finnish capital, there are currently no green roofs that are specifically dedicated to agriculture, but a number of organizations and individuals grow vegetables and fruit in planter boxes on rooftops. Currently, in Helsinki, this type of agriculture is not a necessity, however, it is a popular theme that has gained a momentum in the last few years. Helsinki, like most world capitals, is growing and, according to the Finnish Statistics Agency, the trend is to move and stay in the inner city, rather than the rural outskirts (Finnish population and migration statistic, Tilastokeskus 2011). The city is not very dense - 2,841 person per km<sup>2</sup> (Facts about Helsinki 2013), and has many areas of green space, but this does not mean that there is space for community urban agriculture. Gardening in allotment gardens is popular in Helsinki and most of them have a waiting list of several months before the gardening season begins. In fact, urban farming is becoming a cultural trend with increasing number of young residents participating in gardening activities organized by several organizations (Sipari & Lehtonen 2014). Besides, city gardening allotments are not always conveniently located and sometimes only reachable by car for their tenants. City rooftops could

offer a convenient gardening option at the place of residence or work, especially within the inner city. Given the interest in gardening in Helsinki and the limited availability of farmable land in the city, rooftops could be a suitable place for additional community agriculture possibilities (Wortman & Lovell 2014).

## **1.2. Reasoning for the study**

### **1.2.1. Rooftop agriculture**

Rooftop agriculture can be implemented in a number of ways. The simplest of these is gardening in containers, which may vary in size, shape and materials. Essentially, anything, from plastic swimming pools to grocery bags can be used as a plant container, as long as it can hold soil and plants. This method is simple, does not require any modifications to the roof (provided the roof load capacity is sufficient) and it is easy to move the plants in case of roof repairs or any other disturbances (Foss et. al. 2011). Another option for rooftop agriculture is on green roofs. This method is more involved and implies that the roof surface is covered with a growing medium for plants. Gardening is possible on green roofs that have a thick enough organic layer for plants to develop. Even though this option is more expensive, it offers additional benefits beyond vegetable production and recreation (Foss et. al. 2011). Green roofs provide additional building insulation, roof surface protection, stormwater retention and offer additional habitat for insects and birds in the built urban environment (USEPA 2008). The third type of rooftop gardening using the method of hydroponics. This is the lightest of all of the options as it is soilless and plants are grown in a special nutrient solution, which can increase growth rates and productivity (Foss et. al. 2011).

Finally, rooftop agriculture can be practiced as a combination of the three methods described above but in enclosed greenhouse structures. Growing plants in an enclosed environment on city roofs provides for prolonged gardening opportunities in harsh climates, protects plants from ambient urban pollution, and

recycles energy by harvesting excessive heat escaping from buildings (Foss et. al. 2011).

An experimental design of a green house that harvests excess building heat already exists in Helsinki at the Exactum Building of the Helsinki University (Fig. 1). One of the projects of this green house is the production of chili peppers, which seems to be a successful enterprise that supplies the local university cafeteria with spicy chilies almost year round (Pervilä et al. 2012). The potential of rooftop agriculture is immense if taking into consideration all available rooftop space that could be used for food production. For example, according to Ted Caplow (the executive director of the New York Sun Works company specializing in energy efficient urban green houses) if New York City used all of its roof space for vegetable production, it could provide twice the volume of necessary produce for its residents (Vogel 2009).



**Figure 1.** Exactum greenhouse in Helsinki, Finland during winter and summer 2012. Photographs are taken from opposing directions. Source: Mikko Pervilä

Local food production in cities would not only guarantee access to fresh, nutritious foods to residents (e.g. Minnich, 1983, Duchemin et al., 2008, Leake et al. 2009 Blaine et al. 2010) but also provide opportunities for recreation, exercise and community building (e.g. Patel 1991, Malakoff 1995, Brown & Jameton 2000, Flores 2006, Clayton 2007, Gross & Lane 2007, Leake et al. 2009). Rooftop agriculture could also reduce human impact on the environment (Doron 2005, Flores 2006), however it largely depends on whether it is practiced properly. For example, excessive use of nutrients on rooftops could lead to

leakages from roofs, adding more nutrient pollution to local water bodies (e.g. Berndtsson et al. 2006, Berghage et al. 2009, Razzaghmanesh et al. 2014).

### **1.2.2. Urban environmental conditions and urban farming.**

Since the conditions of urban agriculture differ from the conventional way of growing crops, it poses a question of whether the quality of the produce also differs in its content of nutrients and pollutants. Urban ecosystems have altered atmospheric and microclimatic condition, due to multiple anthropogenic activities. Temperature, humidity, wind, CO<sub>2</sub> and ozone levels are different in cities compared to rural areas, all of which influences plant development (Wortman & Lovell 2014).

Health risks associated with exposure to increased levels of environmental pollutants, such as heavy metals and polycyclic aromatic hydrocarbons, are also the reasons why the mentioned benefits of urban agriculture are often undermined (Alloway 2004, Clark et al. 2006). Increasingly, health issues associated with traffic related urban pollution are gaining attention with policy makers and legislators, which also brings more attention to the question of urban agriculture (WHO 2006, UNEP 2007).

Urban environments are generally more polluted than rural areas and urban crops may be exposed to higher levels of contaminants, including heavy metals and PAHs, due to traffic emissions and combustion of fossil fuels for energy and industrial purposes (e.g., Chaney et al. 1984, Mielke et al. 2011). Experiments have shown that plant contamination with these elements is dependent on many factors such as soil and air quality, the type of species, as well as wind conditions and proximity to pollution sources, such as motor traffic, energy production or industry (Forman & Alexander 1998, Säumel et al. 2012). Heavy metals and PAHs accumulate in urban soils and other surfaces, such as plant shoots and leaves. This causes concerns about growing crops in cities,



which are often more contaminated with these elements compared to rural areas (Smit et al. 1996).

Urban agriculture is especially questionable because there is empirical evidence demonstrating that consumption of vegetables produced on polluted sites can be a cause of serious health problems (e.g. Qadir et al. 2000, Hough et al. 2004, Finster et al. 2004, Clark et al. 2006, Kachenko & Singh 2006, Pruvot et al. 2006, Sharma et al. 2007, Khan et al. 2008). Heavy metals can be toxic to humans and health effects are numerous and may range from skin irritation to organ damage to death depending on the metal, the degree of exposure and metal concentration (Martin & Griswold 2009). PAHs are also harmful and, in fact, they comprise the largest class of chemical compounds that cause cancer (Srogi 2007).

### **1.2.3. Trace elements**

Trace elements, however, are not all toxic or harmful. On the contrary, many metals are essential for the proper functioning of the human body. For example, iron is a crucial part of hemoglobin present in blood (Underwood, 2012). Metals such as zinc (Zn), copper (Cu), manganese (Mn), selenium (Se), chromium (Cr), and molybdenum (Mo) are all trace elements or nutrients, which are important in the human diet. Borderline elements that have no benefit to the human physiology are the ones that are considered toxic, and the main four are lead (Pb), mercury (Hg), cadmium (Cd) and tin (Sn). These elements can cause a threat to human health if taken in quantities above suggested safety limits (EC 2006). Nonetheless, a number of metals (e.g. Cd, Cr, Cu, Mn, Ni, Zn, and Pb) are found in elevated concentrations in urban environments and are linked to traffic and industrial activities (Deletic & Orr 2005, Adedeji et. al 2013). For example, Cu, Zn, Cs and Pb can be originated from rubber tire wear, lubricating oils and gasoline combustion (Shi et al. 2012). The most consistent heavy metal found in high concentrations in cities is Pb, which for many years was added to gasoline. Even though it has been removed from gasoline content more than thirty years

ago, this metal is still persistently present in the environment and has become a long-term environmental issue throughout the world (Shen et. al. 2002).

Human exposure to metals may occur through continuous ingestions in regular diet, medication and contaminated soil; through contact with contaminants in industrial settings; and through exposure to ambient environmental conditions (Inoue 2013). If urban agriculture is not practiced according to suggested safety guidelines (USEPA 2011), trace elements could be present in excessive concentrations in harvested crops. In a study by Säumel et al., (2012) on inner city gardens in Berlin, more than half of all plant samples exceeded EU safety limits for trace metals in food (EC 2006), which underlines potential dangers of urban agriculture.

There are a number of ways to minimize chances for exposure to trace metals in urban plants. The US Environmental Protection Agency recommends using safe gardening practices such as avoiding urban soils and utilizing raised beds with garden mix soil, planting away from roads and other sources of pollution, and choosing plants that are not considered “hyper accumulators” of pollutants of concern (USEPA 2011). For example, according to some studies, fruit producing plants and legumes are low accumulators of trace metals, root vegetables accumulate at moderate rates and leafy green vegetables are high accumulators (Kloke et al. 1984, Ge et al. 2000, Finster et al. 2004, Alexander et al. 2006). Therefore, fruit plants, legumes and root vegetables are more suited for growing in urban setting to minimize possibilities of harvesting crops with increased contaminants concentrations.

#### **1.2.4. Polycyclic aromatic hydrocarbons (PAHs)**

PAHs are a large group of organic pollutants that are toxic, mutagenic and carcinogenic, which is why they cause environmental and health concerns (Weissenfels 1992, Srogi 2007). PAHs can exist not only in particle, but also in gaseous phase, and are transported by air movement (De Nikola et al. 2008).

These compounds naturally occur in the environment and are often a product of forest fires, volcanic eruptions and can be found in coal tar and crude oil (Blumer & Youngblood 1975, Slezakova et al. 2013). They are emitted to the atmosphere as a result of incomplete combustion of biomass or fossil fuels (Desalme et al. 2013). Anthropogenic activities, however, are the major source of contribution of the most dangerous PAHs to the atmosphere. In urban environments they include aerial, water and terrain traffic, energy generation, waste incineration, heating and cooling, and many industrial processes (e.g. oil refining and asphalt manufacturing). Once PAHs are deposited from the atmosphere into soil, they can bind to soil particles, accumulate in plants, leach into ground water, become volatile or eventually degrade (Mumtaz & George 1995).

Studies indicate that human exposure to PAHs occurs primarily through ingestion or inhalation of contaminated soil and ingestion of contaminated food (e.g. Wild et al. 1992, Vyskocil et al. 2000, Cocco et al. 2007), which is why it is crucial to examine PAH concentrations in urban produce. Research suggests that vascular plant contamination occurs both, by direct (air-leaf) and indirect (air-soil-root) pathways, with the direct pathway being the main contributor when plants are grown on non-contaminated soils (Kipopoulou et. al 1999, Simonich & Hites 1994, Lehndorff & Schwark 2004). In other words, plants tissue exposed to ambient conditions can absorb PAHs in the gaseous form and plant roots can uptake PAHs from the soil. Moreover, once the pollutant is in the plant tissue, it can migrate from shoot to root and vice versa (Desalme et. al 2011). This way, plants open up a transfer route for PAHs to reach higher trophic levels like humans, who may consume plants containing the contaminant.

In light of health concerns related to PAHs, which are abundant in urban areas, it is of increased importance to monitor their levels especially in edible plants grown in cities.

### **1.3. Hypothesis**

To my knowledge, no scientific studies exist that have examined levels of pollutants in plants grown on urban rooftops. In light of the increasing popularity of this form of urban agriculture, and due to concerns regarding the quality and safety of edible plants cultivated on urban rooftops, I conducted an experiment in Helsinki, Finland where I grew vegetables on city rooftops and then tested them for possible contamination. Additionally, vegetables were examined washed and unwashed to determine the significance of atmospheric deposition on the surfaces of vegetables in rooftop gardens in the city of Helsinki. I focused on testing trace elements and polycyclic aromatic hydrocarbons in the edible parts of the vegetable tissue, as these directly determine the health risks of consuming products grown on urban rooftops.

The main hypotheses and predictions of this study are the following:

- 1) Urban rooftop vegetables in Helsinki, Finland will have higher concentrations of trace elements and PAHs compared to vegetables available at grocery stores and markets. Due to urban pollution, concentrations of trace elements and PAHs in rooftop vegetables are expected to be higher than the official European safety limits for human consumption.
- 2) Concentrations of atmospheric deposition of contaminants in vegetables grown on rooftops in Helsinki, Finland will reduce after washing.

### **2. Material and methods**

This experiment was a multistep process that involved planting crops, chemical analysis of samples in a laboratory and statistical processing of results. The practical part of the study was done between June and August of 2013.

## 2.1. Experimental design, sample collection and treatments

This study was performed in the city of Helsinki, Finland, however, the experimental design was developed to be easily replicated in any urban center. The study area included five rooftop gardens, located in different parts of the city (Fig. 2), and the experimental procedures were carried out as similarly as possible at all of them. The rooftops were considered replicates in the design of the experiment.

All five locations were supplied with the same basic gardening soil from Kekkilä Oy, organic vegetable seeds and were irrigated with tap water and ambient precipitation. The composition of the soil was 50 % brown peat, 30% black peat, 10% fine sand, 10% compost. The fertilizers in the mix were basic peat fertilizer (NPK 15-10-16) 1.20 kg/m<sup>3</sup> and fine Mg lime 10.00 kg/m<sup>3</sup>. The soil pH was 5.9 and conductivity - 23 mS/m.

Three types of edible species (legume, root and leafy green) were selected for the experiment in order to test contamination levels in plants with different routes (air-leaf, soil-root) and rates (low, medium and high) of contaminant accumulation (Kloke et al. 1984, Ge et al. 2000, Finster et al. 2004, Alexander et al. 2006). One wooden planter box per rooftop was used to grow the vegetables. The three species were planted in the same box and no additional fertilization was applied at any of the locations throughout the length of the experiment. On June 1<sup>st</sup> and 2<sup>nd</sup>, 2013, lettuce (*Lactuca Sativa* “Black Seeded Simpson”), radish (*Raphanus Sativus* “Cherry Belle”) and pea (*Pisum sativum* “Early Onward”) seeds were planted into planter boxes on all five rooftops. Radish and lettuce seeds were planted in lines and when seedlings were about 3-4 cm, the rows were thinned leaving the strongest plants at the distance of about 4-5 cm apart. Pea seeds were also planted in a row, approximately 5-6 cm apart. The vegetables were harvested at maturity. Radish was harvested in three weeks, lettuce in eight, and peas in nine.



**Figure 2.** The five rooftops investigated included (A) the Nokia House head quarter building in Espoo (60.172022 N, 24.827317 E), (B) Kaapeli Center in Ruoholahti (60.161892 N, 24.90626 E), (C) Savoy Restaurant at the city center (60.167091 N, 24.947266 E), (D) HAPPI Youth Center in Kalasatama (60.185564 N, 24.966771 E) and (E) the Environmental Building at the Viikki campus of Helsinki University (60.225429 N, 25.016209 E).

Commercially grown vegetable available at food store and markets were taken as controls. All control samples were acquired from different city grocery stores and farmers' markets, and were all produced by different growers from southern Finland. All control lettuce plants were produced in enclosed greenhouse systems, and peas and radish were grown in open fields. Control samples were acquired and tested in June 2013.

Experimental and control samples were collected into new plastic bags and transported to the laboratory for analysis. Each of the rooftop samples was divided in half so that one half could be tested unwashed and the other washed. All control samples were tested washed. Radish was not tested unwashed, as only the roots are edible and they are always washed before ingestion. To summarize, there were five control samples of each crop washed, five rooftop samples of each crop washed, and five samples of peas and lettuce unwashed. Pea fruit were

tested together with the pods, as they are also edible and are often consumed with the actual peas.



**Figure 3.** Experimental planter box at Kaapeli Center (top left); Container rooftop garden at the Kaapeli Center (top right and bottom left); Rooftop garden at Nokia House (bottom right).

All samples to be tested washed, were rinsed first with tap water and then with ultra purified 18.2 M $\Omega$  (mega ohms) water. To prepare samples for analysis, they were first disintegrated using the Ultra-Turrax homogenizer. Then, the homogenized sample mass was transferred into HNO<sub>3</sub>-washed glass containers, frozen, and then dehydrated in a freeze-dryer (Fig. 3B). Once the samples were dry, two measurements of approximately 0.5 grams were taken, one for the heavy metal analysis and the other for PAH analysis (Fig. 3A).

## 2.2. Trace elements analysis

Concentrations of trace elements (aluminum (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), zinc (Zn), lead (Pb), arsenic (As) and vanadium (V)) in samples were analyzed using the

Inductively Coupled Plasma Mass Spectrometry device (ICP-MS). In preparation for the ICP-MS analysis, dry vegetable samples were first homogenized with an Ultraturrax mixer, dried in a freeze-drier and then processed in a Microwave Accelerated Reaction System 6 (MARS, by CEM Corporation, USA) by being dissolved in 10 mL of concentrated Nitric acid and run through a “Plant Material” cycle in MARS (Fig. 3C). Then, the samples were diluted with ultra pure water to 2% nitric acid concentration (v/v), and 5mL of each sample transferred into testing tubes. 50  $\mu$ L of indium (1000  $\mu$ g/L) internal standard (In) was added to each sample and finally processed in the Elan 6000 (by Perkin-Elmer) ICP-MS device. One blank sample was prepared for every batch of samples following the same procedures as the actual samples. Additionally, two ICP control samples, 100 and 5  $\mu$ g/L, were prepared from 100  $\mu$ g mL solution (Roth XIII). All standards, blanks and controls were in the same nitric acid concentration as the samples. The calibration was done with a series of standards and using linear regression to obtain a calibration line for each element. The calibration mix used in the analysis was Mixture XIII from Merck. The control solution was acquired from VWR International, Leuven, Belgium.

### **2.3. Polycyclic aromatic hydrocarbons (PAHs) analysis**

PAH concentrations in samples were measured by the gas chromatograph-mass spectrometer (GC-MS) instrument. The analysis was done for 16 compounds (naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FL), phenanthrene (PHEN), anthracene (ANT), fluoranthene (FLR), pyrene (PYR), benzo(a)anthracene (B(a)A), chrysene (CHY), benzo(b)fluoranthene (B(b)F), benzo[k]fluoranthene (B(k)F), benzo(a)pyrene (B(a)P), indeno(1,2,3-cd)pyrene (INP), dibenzo(a,h)anthracene (DBA), benzo(ghi)perylene (BPY)).

Sample preparation for the GC-MS analysis involved several steps. Disintegrated and freeze-dried plant material (approximately 0.50 g) was transferred into test tubes and weighed. A mixture of deuterated PAHs (50  $\mu$ L, 4



ng  $\mu\text{L}$ ) was added as internal standards to the tubes followed by 15 mL of 1:1 (v:v) acetone-hexane solvent mixture. Then, the samples were sonicated for 30 min and placed in a shaker (200 rpm) overnight. After that, the sample extracts were transferred to test tubes and evaporated almost to dryness with a gentle nitrogen stream. Approximately 1 mL of hexane was added to sample test tubes and purified with silica gel columns. Approximately 1 g of activated (160 °C overnight) silica gel was placed into Pasteur-pipettes fortified with cotton wool plugs, and rinsed with hexane (3 mL). Then, the samples were added to columns and PAH-compounds were eluted with 10 mL dichloromethane (DCM) (Fig.3D). After that, samples were concentrated with nitrogen stream to 0.5 mL volume and the remaining samples were transferred into analytical vials. Before the final analysis by GC-MS 20  $\mu\text{L}$ , 10 ng  $\mu\text{L}$  of deuterated anthracene was added to each sample vial as a recovery standard. One blank sample was prepared with each sample batch following the same procedures of the actual samples described above.



**Figure 4.** Laboratory analysis procedures. Top left – measuring pretreated crop samples for trace element analysis. Top right – crop samples after they have been disintegrated and freeze-dried. Bottom left – preparing samples for the MARS treatment, as part of trace element analysis. Bottom right – crop sample extract eluted with dichloromethane, as part of the PAH analysis.

The analysis was performed in the Shimadzu GC-MS-QP2010 Ultra system equipped with the AOC-20i/AOC-20s autosampler (Corp., Kyoto, Japan). The internal standard mix for this method (PAH-Mix 31 deuterated) contained naphthalene-D8, acenaphthene-D10, phenanthrene-D10, chrysene-D12 and perylene-D12. It was purchased from Dr. Ehrenstorfer GmbH. Deuterated anthracene-D10 was also purchased from the same place. The solvents (acetone, hexane and dichloromethane) were of liquid chromatography (LC) quality and were acquired from Merck KGaA, Darmstadt, Germany. Silica gel 60 was also purchased from Merck KGaA.

The recoveries of internal standards were acceptable for the standards (acenaphthene-D10  $35\pm 12\%$ , phenanthrene-D10  $41\pm 9\%$ , chrysene-D12  $49\pm 11\%$ , perylene-D12  $77\pm 20\%$ ) except for naphthalene-D8 ( $15\pm 11\%$ ), which was too low, making results for this compound unreliable.

#### **2.4. Statistical analyses**

Trace element and PAH data that were not normally distributed were transformed (either logarithmically or square-root) to approximate normality. If the data were normal (either with or without transformations), a *t*-test was used for statistical comparisons. A Wilcoxon signed-rank test was applied for non-parametric data. These tests were used to evaluate differences in trace elements and PAHs between roof garden vegetables, and vegetables obtained from grocery stores or farmers' markets. For the comparison of washed and unwashed roof samples, a paired version of the tests was used, since each set of washed and unwashed samples came from the same plant/batch of plants. All figures were constructed using the raw data. For elements where more than two samples showed results below limits of quantification in each treatment, statistical analysis was not possible due to insufficient data.

### 3. Results

#### 3.1. Concentrations of trace elements and PAHs in roof versus control samples

Generally, the content of trace metals and PAHs differed among plant species of both treatments – roof garden samples and control samples (Table 1). A number of samples had concentrations of trace elements and PAHs below quantification limits.

**Table 1.** Concentrations of trace elements in vegetables grown on rooftop gardens in Helsinki, Finland and in vegetables from stores [mg/kg dw]: median (Med), minimum (Min), maximum (Max); 5 indicates the number of samples; <LOQ indicates values below limit of quantification.

Trace element content in [mg/kg dw]		Species / Source					
		Peas		Lettuce		Radish	
		Roof (5)	Control (5)	Roof (5)	Control (5)	Roof (5)	Control (5)
Aluminum (Al)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	<LOQ	35	<LOQ	<LOQ	53
Cadmium (Cd)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.10
	Max	<LOQ	<LOQ	0.07	<LOQ	<LOQ	0.31
Cobalt (Co)	Min	<LOQ	0.03	<LOQ	0.05	<LOQ	<LOQ
	Med	<LOQ	0.04	<LOQ	0.13	<LOQ	0.05
	Max	<LOQ	0.07	<LOQ	2.5	<LOQ	0.25
Cromium (Cr)	Min	<LOQ	0.30	<LOQ	4.3	<LOQ	<LOQ
	Med	<LOQ	0.35	<LOQ	8.5	<LOQ	2.6
	Max	<LOQ	0.46	4.5	180	1.7	37
Copper (Cu)	Min	1.1	3.9	2.3	2.1	<LOQ	2.0
	Med	1.3	4.2	3.9	3.0	<LOQ	3.9
	Max	3.2	5.1	4.5	7.1	<LOQ	4.5
Manganese (Mn)	Min	4.5	5.5	27	31	6.3	2.3
	Med	6.2	6.5	54	67	7.7	7.7
	Max	31	8.5	130	130	9.9	9.8
Nickel (Ni)	Min	<LOQ	<LOQ	<LOQ	1.6	<LOQ	<LOQ
	Med	<LOQ	0.79	<LOQ	5.0	<LOQ	<LOQ
	Max	<LOQ	0.88	3.2	120	<LOQ	11
Zinc (Zn)	Min	15	<LOQ	16	22	12	14
	Med	20	22	22	44	14	27
	Max	28	24	43	50	16	33
Lead (Pb)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	<LOQ	<LOQ	1.5	0.52	<LOQ
Arsenic (As)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
Vanadium (V)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ

**Table 2.** Content of PAHs in vegetables grown in rooftop gardens in Helsinki, Finland and in vegetables from stores [ng/g dw]: median (Med), minimum (Min), maximum (Max); 5 indicates the number of samples; <LOQ indicates values below limit of quantification.

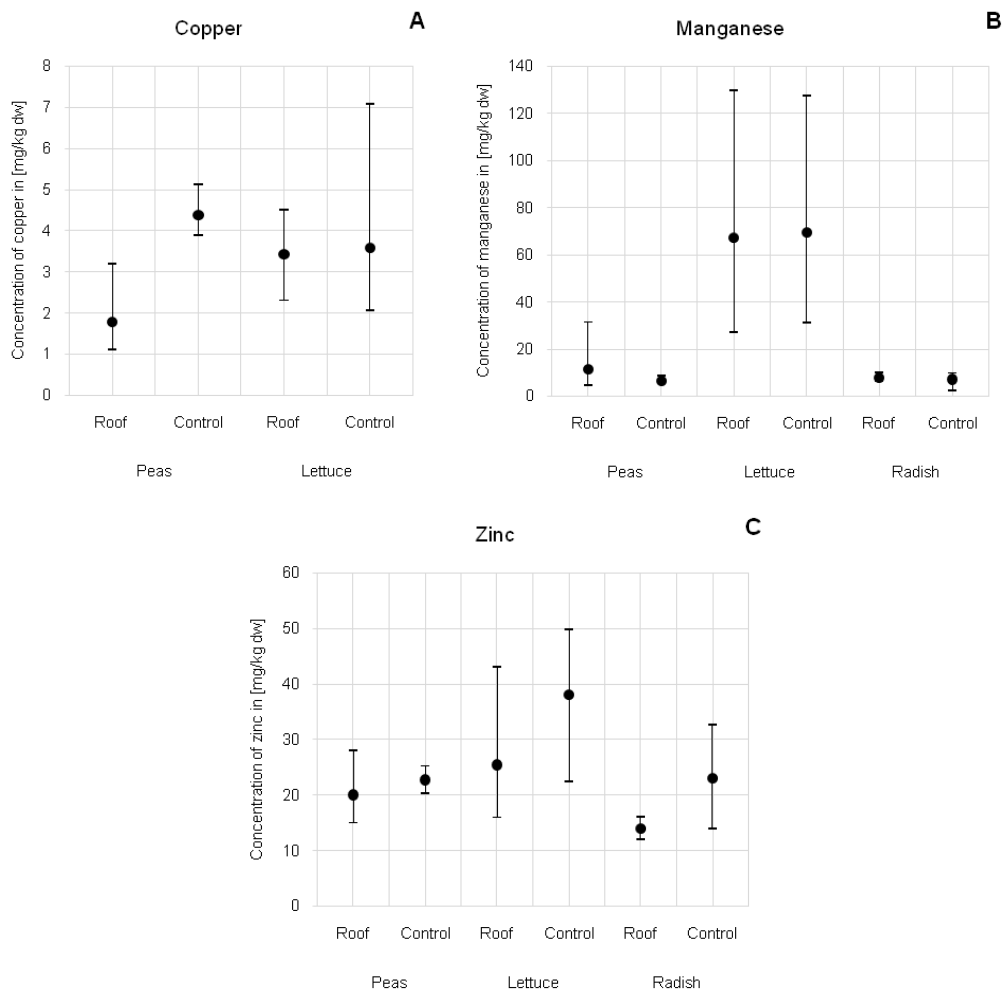
PAH compounds in [ng/g dw]		Species / Source					
		Peas		Lettuce		Radish	
		Roof (5)	Control (5)	Roof (5)	Control (5)	Roof (5)	Control (5)
Naphthalene (NAP)	Min	3.9	<LOQ	<LOQ	<LOQ	<LOQ	13
	Med	6.9	<LOQ	8.9	20	31	44
	Max	14.0	6.9	82	24	82	90
Acenaphthylene (ACY)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	1.1	1.9	<LOQ	0.89
	Max	<LOQ	1.5	2.2	2.7	1.1	2.4
Acenaphthene (ACE)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	1.1	<LOQ	<LOQ	<LOQ	1.6	<LOQ
Fluorene (FL)	Min	1.6	1.4	<LOQ	2.9	2.0	<LOQ
	Med	1.9	2.4	4.8	8.1	3.3	3.4
	Max	2.4	3.0	8.0	8.9	4.4	4.8
Phenanthrene (PHEN)	Min	7.4	14	6.8	33	10	12
	Med	9.8	16	54	79	17	26
	Max	24	67	110	93	35	47
Anthracene (ANT)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	1.4	1.5	<LOQ	0.60	1.2
	Max	0.90	3.4	4.6	4.3	2.0	2.8
Fluoranthene (FLR)	Min	<LOQ	5.9	2.2	14	3.6	5.1
	Med	<LOQ	7.8	13	20	7.2	6.8
	Max	1.5	23	44	31	11	19
Pyrene (PYR)	Min	3.6	11	5.4	12	4.6	9.7
	Med	4.1	14	23	24	8.0	12
	Max	6.7	29	55	67	33	25
Benzo[a]anthracene (B(a)A)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.68
	Max	<LOQ	1.5	9.9	<LOQ	1.3	3.9
Chrysene (CHY)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	0.57	<LOQ	<LOQ	1.2	0.34
	Max	<LOQ	5.5	7.0	<LOQ	4.2	3.7
Benzo[b]fluoranthene (B(b)F)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	0.83	<LOQ	<LOQ	<LOQ	0.51	<LOQ
	Max	1.6	<LOQ	1.4	<LOQ	1.7	<LOQ
Benzo[k]fluoranthene (B(k)F)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	0.47	<LOQ
	Max	<LOQ	<LOQ	1.0	3.1	1.6	<LOQ
Benzo[a]pyrene (B(a)P)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	1.4	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	15	0.06	<LOQ	1.4	<LOQ
Indeno[1,2,3-cd]pyrene (INP)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	1.8	<LOQ	<LOQ	1.5	1.4
Dibenzo[a,h]anthracene (DBA)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
Benzo[ghi]perylene (BPY)	Min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Med	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
	Max	<LOQ	1.6	<LOQ	4.8	4.5	0.56

Since many samples showed results below quantification limits, I was able to perform statistical analysis and make comparisons only for some elements (copper, manganese, and zinc) and compounds (acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, and chrysene). The difference between roof and control samples was insignificant ( $p > 0.050$ ), except for copper and pyrene in peas, which showed significant results (Table 3). Also, phenanthrene in peas and zinc in lettuce and radish had results close to statistical importance (Table 3). This demonstrates that there is, largely, no significant difference between roof and store samples, regarding concentrations of trace elements and PAHs.

**Table 3.** T-test and Wilcoxon signed rank test results for comparison of roof and control samples of peas, lettuce and radish. Mean and standard deviations are also presented. \* = t-test, \*\* = Wilcoxon signed rank test. Concentrations of trace elements in [mg/kg dw] and PAHs [ng/g dw].

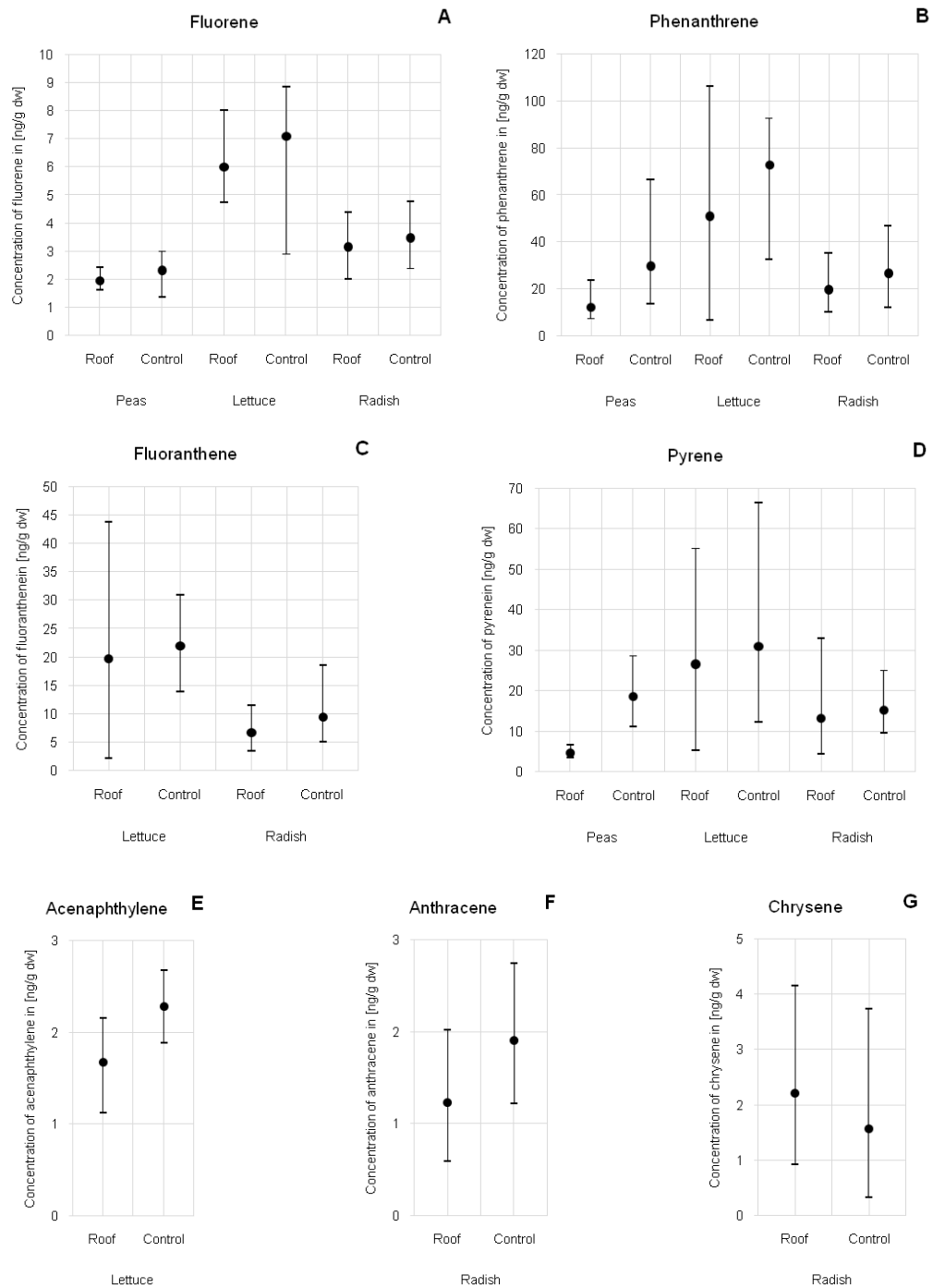
Plant & Element	Roof	Control	Test statistic (W/t-value)	P-value
	Mean (SD)	Mean (SD)	(W/t)	
<b>Peas</b>				
Copper (Cu)**	1.8 (0.89)	4.4 (0.50)	0.0	0.0079
Manganese (Mn)**	12 (11)	6.6 (1.2)	12	1.0
Zinc (Zn)*	20 (5.0)	23 (2.2)	-1.2	0.29
Fluorene (Fl)*	2.0 (0.31)	2.3 (0.61)	-1.2	0.27
Phenanthrene (Phen)**	12 (6.5)	30 (23)	3.0	0.056
Pyrene (Pyr)**	4.7 (1.3)	19 (8.7)	0.0	0.0079
<b>Lettuce</b>				
Copper (Cu)**	3.4 (1.0)	3.6 (2.0)	10	1.0
Manganese (Mn)*	67 (41)	69 (37)	-0.81	0.45
Zinc (Zn)**	25(10)	38 (13)	2.0	0.063
Acenaphthylene (Acy)*	1.7 (0.52)	2.3 (0.40)	-1.6	0.20
Fluorene (Fl)**	6.0 (1.6)	7.1 (2.4)	5.0	0.29
Phenanthrene (Phen)**	51 (38)	73 (25)	7.0	0.31
Fluoranthene (Flr)**	20 (17)	22 (6.4)	9.0	0.55
Pyrene (Pyr)**	27 (19)	31 (21)	11	0.84
<b>Radish</b>				
Manganese (Mn)**	7.8 (1.4)	7.1 (3.0)	14	0.84
Zinc (Zn)**	14 (1.4)	23 (8.2)	3.5	0.067
Fluorene (Fl)**	3.2 (1.0)	3.5 (0.98)	7.0	0.56
Phenanthrene (Phen)**	20 (9.5)	27 (14)	10	0.69
Anthracene (Ant)*	1.2 (0.72)	1.9 (0.78)	-1.2	0.32
Fluoranthene (Flr)*	6.8 (3.3)	9.5 (5.7)	-0.91	0.39
Pyrene (Pyr)**	13 (11)	15 (6.8)	8.0	0.42
Chrysene (Chy)**	2.2 (1.5)	1.6 (1.9)	9.0	0.40

Contrary to expectations, concentrations of trace metals and PAHs were higher in control samples, rather than in roof samples. The concentration of copper was 2.45 times higher in control samples in peas, in lettuce the results were similar, and in radish, copper was below quantification limit in roof samples (Table 3, Fig. 5A). Manganese concentration was similar in roof samples and controls in lettuce and radish, but in peas roof samples, Mn concentration was 1.75 times higher than in controls (Table 3, Fig. 5B). Zinc concentration was higher in controls for all three crops by 1.13 times in peas, 1.49 times in lettuce and 1.65 times in radish (Table 3, Fig. 5C).



**Figure 5.** Concentrations of trace elements (Cu, Mn and Zn) in rooftop crops versus control crops in [mg/kg dw]. Graph A represents concentrations of copper, graph B concentrations of manganese and graph C concentrations of zinc. For statistical test results, see Table 3.

The concentration of the PAH fluorene was higher in control samples in peas by 1.19 and in lettuce by 1.18 times; however, in radish, fluorene concentrations were similar between roof and control samples (Table 3, Fig. 6A). Phenanthrene concentration was higher in control samples for all three crops by 2.41 times in peas, 1.43 times in lettuce and 1.35 times in radish (Table 3, Fig. 6B). Fluoranthene concentration was also higher in controls by 1.11 times in lettuce and 1.40 times in radish, but in peas it was below the quantification limit (Table 3, Fig. 6C). Pyrene concentration was also higher in control samples for all three crops by 3.96 times in peas, 1.16 in lettuce and 1.16 in radish (Table 3, Fig. 6D). Acenaphthene level was 1.36 times higher in lettuce controls compared to roof samples, and it was below the quantification limit in peas and radish (Table 3, Fig. 6E). Anthracene level was 1.54 times higher in controls in radish, but in peas and lettuce, it was below quantification (Table 3, Fig. 6F). Chrysene was below quantification limit in peas and lettuce, but in radish, its concentration was higher in the roof samples by 1.40 times (Table 3, Fig. 6G).



**Figure 6.** Concentrations of PAHs in rooftop crops versus control crops in [ng/g dw]. Graph A represents concentrations of fluorene, graph B concentrations of phenanthrene, graph C concentrations of fluoranthene, graph D concentrations of pyrene, graph E concentrations of acenaphthylene, graph F concentrations of anthracene and graph G concentrations of chrysene. For statistical test results, see Table 3.



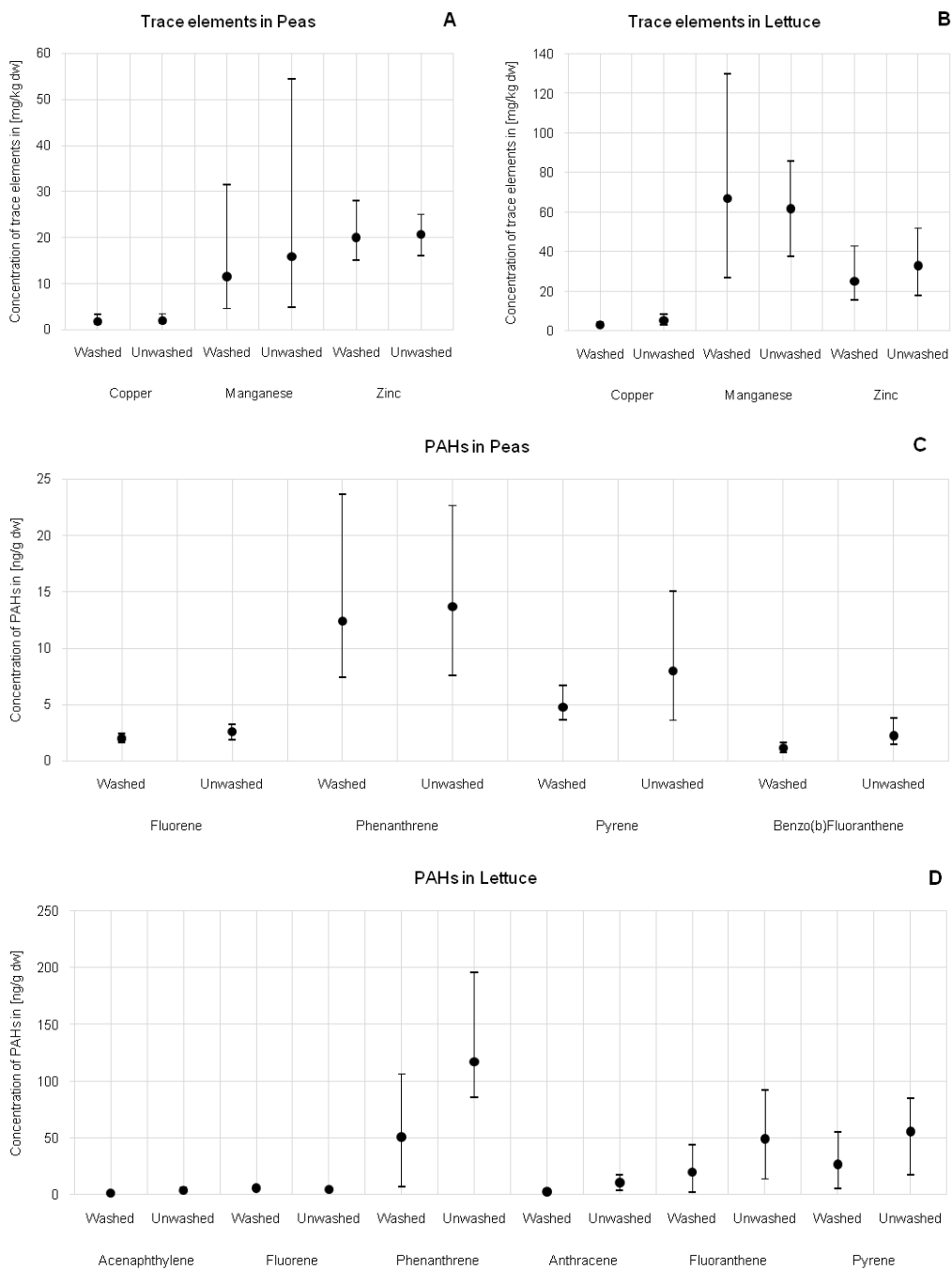
### 3.2. Comparison of washed and unwashed sample concentrations

The results of the comparison of roof samples washed and unwashed demonstrated a trend of concentrations decreasing after washing in most cases, as expected (Table 4, Fig. 7).

**Table 4.** Paired t-test and Paired Wilcoxon signed rank test results for comparison of washed and unwashed roof samples of peas and lettuce. Means and standard deviations are also presented. \* = Paired t-test, \*\* = Paired Wilcoxon signed rank test. Concentrations of trace elements in [mg/kg dw] and PAHs [ng/g dw].

Plant & Element	Washed	Unwashed	Test statistic	P-value
	Mean (SD)	Mean (SD)	(W/t-value) (V/t)	
<b>Peas</b>				
Copper (Cu)	1.8 (0.89)	2.0 (0.87)	7.0	0.58
Manganese (Mn)	12 (11)	16 (22)	8.0	1.0
Zinc (Zn)*	20 (5.0)	21 (3.7)	0.77	0.48
Fluorene (Fl)	2.0 (0.31)	2.6 (0.53)	13	0.19
Phenanthrene (Phen)	12 (6.5)	14 (6.4)	7.0	1.0
Pyrene (Pyr)	4.7 (1.3)	8.0 (4.5)	14	0.13
Benzo[b]fluoranthene (B(b+k)F)*	1.1 (0.43)	2.2 (1.1)	3.3	0.046
<b>Lettuce</b>				
Copper (Cu)*	3.4 (1.0)	5.5 (2.3)	1.7	0.19
Manganese (Mn)	67 (41)	62 (20)	10	0.098
Zinc (Zn)	26 (10)	33 (14)	10	0.098
Acenaphthylene (Ace)*	1.7 (0.52)	4.1 (2.2)	1.6	0.25
Fluorene (Fl)	6.0 (1.6)	4.9 (1.4)	1.0	0.25
Phenanthrene (Phen)*	51 (38)	120 (45)	2.8	0.048
Anthracene (Ant)*	2.9 (1.6)	11 (6.8)	2.1	0.17
Fluoranthene (Flr)	20 (17)	49 (29)	15	0.063
Pyrene (Pyr)	27 (19)	56 (26)	14	0.13

In pea samples, washing reduced concentrations of copper by 10%, manganese by 27% and zinc by 3% (Table 4, Fig. 7A). PAH concentrations were also reduced – fluorene by 24%, phenanthrene by 9%, pyrene by 41%, benzo(b)fluoranthene by 49% (Table 4, Fig. 7C). In lettuce, washing reduced the concentration of copper by 38% and zinc by 23%. However, concentrations of manganese were higher after washing by 8% (Table 4, Fig. 7B). PAH concentrations also decreased in acenaphthene by 59%, phenanthrene by 56%, anthracene by 73%, fluoranthene by 60% and pyrene by 52%. Concentrations of fluorene, however, increased after washing by 19% (Table 4, Fig. 7D).



**Figure 7.** Concentrations of trace metals [mg/kg dw] and PAHs [ng/g dw] in washed samples versus unwashed samples. Graph A and B show the comparison of contents of trace elements in peas and lettuce respectively, while graphs C and D show the comparison of contents of PAHs in the same samples respectively.

The majority of results were, however, statistically insignificant with two exceptions - benzo(b)fluoranthene in peas, and phenanthrene in lettuce (Table 4).

Manganese, zinc and fluoranthene in lettuce had results close to statistical significance of  $p < 0.050$  (Table 4). The differences between concentrations of these trace metals and PAHs in samples decreased significantly after washing.

#### **4. Discussion**

In light of the rising popularity of urban rooftop farming throughout the world, it is important to take into consideration urban contamination as one of the health risk factors. However, it is also important to remember that environmental conditions vary from city to city, which in turn affects urban crops. For example, PM<sub>10</sub> measurement for Helsinki in 1999 was 19  $\mu\text{g}/\text{m}^3$ , while for New York it was 24  $\mu\text{g}/\text{m}^3$  and for Rome 52  $\mu\text{g}/\text{m}^3$  (Baldasano et al. 2003). Depending on levels of ambient contamination in a particular city, concentrations of contaminants in urban crops may differ greatly (e.g. Sanchez-Camazano et al. 1994, De Nicola et al. 2008, Nabulo et al. 2012, Säumel et al. 2012).

Crops grown on rooftops in Helsinki as part of this study did not show higher concentrations of trace elements and PAHs compared to control samples, which contrasts my original hypothesis (1), according to which the concentrations were expected to be higher in rooftop samples. Additionally, the concentrations were well below safety standards established in the European Union (EC 2006) demonstrating that these rooftop crops were safe enough for human consumption (Table 1, Table 2). On the other hand, washed rooftop samples had lower concentrations of contaminants (trace elements and PAHs) compared to the unwashed rooftop samples, supporting my hypothesis (2). This demonstrated that a significant part of ambient contaminants deposited on plant surfaces can be removed by mechanical washing, as established in other studies (e.g. Ward et al. 1977, Nabulo et al. 2012, Ugolini et al. 2013).

#### **4.1. Factors that may have resulted in low concentrations**

Contaminants in soil may be one of the main risks factors for the production of healthy crops in urban environments (Wortman & Lovell 2014). In this study, the quality of soil was insured by planting the crops into uncontaminated new gardening soil, which is one of the advised best management practices in urban agriculture (USEPA 2011). The contaminants that could have appeared in samples would have come mainly from atmospheric deposition. Since one of the major contributors to increased heavy metal and PAH concentrations in cities is traffic, the most contaminated areas are typically along roadside where contamination accumulates in soil and is also suspended in the air in the form of aerosols. Motorized traffic causes turbulence resulting in atmospheric suspension of some soil particles contaminated with heavy metals and PAHs, which can then settle on the surfaces of plant tissue (Biasioli et al. 2007, Laidlaw et al. 2012).

Nonetheless, even though all sites of this study were alongside urban streets with steady traffic, they were all at heights above street level. This could be one of the reasons why trace metal and PAH concentrations in the study samples were negligible, however studies have shown that urban air quality at heights above street level is irregular. The presence of urban canyons in urban built environment causes irregular airflow and vertical mixing which results in unpredictable air quality at height above the street level (Building height fact sheet, 2014). Chrysikou et al. (2009) established in their study in Thessaloniki, Greece that particle concentrations of all sizes were higher at the street level than at the rooftop as a result of more intensive traffic emissions and road dust. However, the results for organic contaminants did not show significant differences between the heights. Jung et al. (2011) who tested levels of PAHs in New York City, also did not see significant changes in concentrations with changing heights. Quang et al. (2012) results from testing in Australia, presented inconsistent results, where in certain sites particle concentrations reduced with heights and in others increased. Since no air quality test were conducted in this

study to measure the difference in contamination at street and rooftop level, it is difficult to judge whether plants were exposed to less contamination on rooftops.

Another reason for low levels of trace elements and compounds in the study samples could be the overall good air quality of the city. NO<sub>2</sub> and CO levels in Helsinki are comparable to other Central European Cities, while SO<sub>2</sub> levels are lower (Kukkonen et al. 1999). According to the Helsinki Region Environmental Services Authority (Helsingin seudun ympäristöpalvelut), air quality in Helsinki is one of the best among metropolitan areas in Europe (Air quality in the Helsinki Metropolitan Area 2007). This, in part, is due to its location and geography. The city is located on relatively flat terrain and is at the sea where wind is constant, all of which facilitates dilution of air contamination, and therefore, improves overall air quality (Air quality in the Helsinki Metropolitan Area 2007). Additionally, the climate in the city is humid continental (Köppen: Dfb) and there are frequent precipitation events (Kottek et al. 2006). Raindrops act as efficient agents for contamination removal, accumulating particles of different sizes and removing them from the atmosphere (Wexler 1961). These removed particles are then deposited onto different surfaces including plants. However, since in this study the plants were tested washed, most of the deposited atmospheric contaminants were mechanically removed. Hence, the overall low levels of contamination in the ambient conditions and washing could be contributing factors to the outcome of the study.

Low levels of contaminants were detected in all three species regardless of their rate of contaminant accumulation (lettuce - high, radish - medium, peas - low). This shows that there is little ambient contamination with trace metals and PAH in Helsinki, at least in the summer time when the samples were grown. Studies indicate that concentrations of PAHs and heavy metals in urban areas tend to be lower in the summer time than in the winter due to fewer emission sources (e.g. Tuominen et al. 1988, De Nicola et al. 2005, Zang & Tao 2008, Odat & Alshammari 2011). For example, a clear temporal trend for PAH concentrations was evident in a study of *Quercus ilex* leaves collected in Naples, with lower concentrations in the summer than in the winter (De Nicola et.al.

2005). Therefore, seasonal changes could also be the reason for negligible contamination levels in samples of this study.

The results of this study showed a clear and constant trend of lower concentrations of trace elements and PAHs in roof samples rather than in control samples and a reduction of contamination after washing the crops. However, it is important to mention that in every chemical analysis there is a margin of uncertainty that should be taken into consideration. In this study with multiple pretreatments, such as disintegration, freeze drying, extraction, dilution and others (see materials and methods section), the margin of uncertainty is around 15%.

#### **4.2. Previous studies and comparison limitations**

Field studies conducted in cities are few and no studies have specifically concentrated on evaluating the quality of urban rooftop horticultural crops. The only similar experiment that I am aware of has been performed in New York, where heavy metal concentrations were measured in samples of rooftop lettuce and compared to those of lettuce produced on a rural farm. The outcome of the experiment showed negligible concentrations of heavy metals in roof samples, which is similar to the results of this study in Helsinki (Arky et al. 2012). No similar studies have been performed previously in the Finnish capital, and this is the first study to focus specifically on urban rooftop crops and examine them for contaminants prominent in cities.

The study most similar to this one, but done at a street level in allotment gardens, was done by Säumel et al. (2012) in Berlin, Germany, however they only tested concentration of trace elements and not PAHs. Other urban studies that I use for reference evaluated contamination levels in crops from contaminated urban allotments, or they tested existing vegetation (trees and grass) either from allotment gardens or other urban green spaces. For these reasons, it is a challenge to make comparisons between my experiment and these studies

because of the array of variables that affect the results (e.g. soil quality, vegetation type, street level vs. rooftop, etc.).

I did not find that crops grown on urban roofs in Helsinki had increased concentrations of trace metals and PAHs, which does not support the general claim that urban agriculture poses a health risk. Overall, the concentrations of trace metals in samples from this experiment were much lower than in the Salamanca, Spain study of urban allotment soil and grass by Sanchez-Camazano et al. (1994), or the Berlin, Germany study by Säumel et al. (2012) that focused on urban crops at ground level. For example, the results for Cd were below quantification limits in all samples except for one lettuce sample 0.07 mg/kg. Similarly, Pb was detected only in one radish sample 0.52 mg/kg. The concentration range of Cd reported in the Salamanca study was 0.57-1.77 mg/kg and Pb was 1.07-10.83 mg/kg in grass from urban gardens. In the Berlin study, Cd concentration had the range of 0.01-1.23 mg/kg and Pb 0.1-32.2 mg/kg in various vegetables from inner city allotments. PAHs also showed results lower than in other studies that tested urban vegetation (De Nicola et al. 2008). For instance, *Quercus ilex* leaves from Naples, Italy had concentrations of benzo(a)pyrene of 47.10 ng/g dw, where mean from the this Helsinki study for all samples was 0.81 ng/g dw (De Nicola et al. 2008).

It was surprising to discover that in many cases contaminant levels were higher in control samples (e.g. Cu in roof peas 1.78 mg/kg vs. control peas 4.36, Phen in roof lettuce 51.05 ng/g vs. control lettuce 72.82 ng/g), especially because these store/market-bought samples, not exposed to the environment of the study, were assumed to be contaminant-free (or with contaminant levels below established safety limits (EC, 2006)). Concentrations were still well below the established safety limits (Table 1, Table 2), but the fact that roof samples had even lower concentrations was unexpected. Säumel's et al. (2012) study that used supermarket vegetables for controls, also revealed presence of minimal levels of contaminants in those samples. The reason for using store/market samples as controls is due to the fact that such produce is officially approved for consumption, is considered to be safe and contaminant-free and is the only other

alternative to self-grown vegetables. The presence of contamination in control samples raises a question of the source of this contamination, which could be due to the quality of air, water and soil, or the use of fertilizers and other agrochemicals. One of the benefits of growing your own crops is that most of these factors (except for air) could be controlled, therefore reducing possibilities for contamination in crops. This study demonstrated that in a relatively clean city like Helsinki, if grown in uncontaminated soil, urban rooftop plants could even be of better quality (in term of contaminants) than commercially produced crops.

### **4.3. The effectiveness of washing to remove contaminants**

The comparison of washed and unwashed roof samples was done to establish the difference between contaminants deposited on the surface of samples and contaminants accumulated within the plant tissue (Alfani et al. 2000). The differences between the treatments showed levels of particle contamination that can be mechanically removed through washing. This also means that the percentage removed by washing is ambient contamination that was deposited on the plant's surface. According to Ward et al. (1977), washing can reduce trace metal concentrations between 10 - 30%, and in my case it was up to 38% (see Results section). This could be due to the fact that the samples were washed twice, first with tap water and then with ultra purified water. Overall, my results support the hypothesis that washing is an effective method to reduce levels of contamination in plants grown on urban rooftops.

Most PAHs showed even higher percentages of reduction of concentrations than trace metals, meaning that a large fraction of PAHs bound to particles can be removed by washing, which contrasts with other studies (e.g. Horstmann & McLachlan 1998, Bakker et al. 2001). According to De Nicola et al. (2008), significantly higher concentrations of PAHs in unwashed versus washed samples only occur for high molecular weight PAHs, as they have higher binding affinities to particulates. In this study, PAHs with light molecular weights, such as phenanthrene and pyrene, also had a considerable reduction in



concentration after washing. The PAH concentration left in washed samples could be partially attributed to PAHs ability to turn into a gaseous form and penetrate plant leaves where they may accumulate (Franzaring 1997, Lehndorff a& Schwark 2004).

#### 4.4. Safety of analyzed samples according to the European Union legislation

Overall, none of the roof or control samples from the study exceeded the European Union threshold value for heavy metal concentrations in food that exist for Pb and Cd (Table 5).

**Table 5.** The limit fixed by the EC Regulation 835/2011 for lead, cadmium. The study results were processed using dry weights (data given in previous tables), but since the EC limits are established in wet weights, roof sample maximum values have been recalculated to facilitate comparison. <LOQ indicates values below limit of quantification of the measuring instrument.

Elements and classification of products		EC Regulation Limit [mg/kg ww]	Roof Samples (Max) [mg/kg ww] (dw)
<b>Lead (Pb)</b>			
	(Peas)	0.20	<LOQ
Legume vegetables, cereals and pulses			
Brassica vegetables, leaf vegetables and the following fungi: Agaricus bisporus (common mush-room), Pleurotus ostreatus (Oyster mushroom), Lentinula edodes (Shiitake mushroom)	(Lettuce)	0.30	<LOQ
Vegetables, excluding brassica vegetables, leaf vegetables, fresh herbs, fungi and seaweed. For potatoes the maximum level applies to peeled potatoes.	(Radish)	0.10	0.026 (0.52)
<b>Cadmium (Cd)</b>			
Vegetables and fruit, excluding leaf vegetables, fresh herbs, leafy brassica, fungi, stem vegetables, root and tuber vegetables and seaweed.	(Peas)	0.050	<LOQ
Leaf vegetables, fresh herbs, leafy brassica, celeriac and the following fungi: Agaricus bisporus (common mushroom), Pleurotus ostreatus (Oyster mushroom), Lentinula edodes (Shiitake mushroom).	(Lettuce)	0.20	0.0028 (0.070)
Stem vegetables, root and tuber vegetables excluding celeriac. For potatoes the maximum level applies to peeled potatoes.	(Radish)	0.10	<LOQ

PAH concentrations were also below the established limits (Table 6). Since the current EU legislation does not establish PAH standards specifically for plants, the comparison of sample results was done using limitations established for processed cereal-based foods and baby foods for infants and young children. According to the product classification from the regulation, the lowest level of PAH presence is prescribed to this category, thus making it the strictest among other mentioned foods (EC 2006). Levels of PAHs are monitored by measuring concentrations of benzo(a)pyrene and/or the sum of 4 PAH compounds (benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene) and are used as markers for the group (EC 2006, Purcaro et al. 2013).

**Table 6.** The limit fixed by the EC Regulation 835/2011 for benzo(a)pyrene and the sum of PAH4 (benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene). The study results were processed using dry weights (data given in previous tables), but since the EC limits are established in wet weights, roof sample maximum values have been recalculated to facilitate comparison.

Classification of products	EC Regulation Limit		Roof Samples (Max)	
	Benzo(a)pyrene [ng/g ww]	Sum of PAH4 [ng/g ww]	Benzo(a)pyrene [ng/g ww] (dw)	Sum of PAH4 [ng/g ww]
<b>Polycyclic aromatic hydrocarbons (PAHs)</b>				
Processed cereal-based foods and baby foods for infants and young children	1.0	1.0	0.068 (1.4)	0.68 (17)

## 5. Conclusion

To summarize, my study demonstrated that edible crops grown on rooftops in Helsinki, Finland have lower concentrations of trace elements and PAHs than in control samples and are safe to consume. I also showed that atmospheric deposition is an important factor when evaluating contamination levels in plants, and mechanical washing is an effective method for contamination reduction. These results are specific to the city of Helsinki and could not be generalized and applied to other urban areas, as air quality and other environmental factors differ from city to city. In light of increasing popularity of

rooftop agriculture throughout the world, further research is needed to establish the safety of this practice, in regards to quality of rooftop horticultural crops.

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