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A study on air quality and heavy metals content of urban food produced in a Mediterranean city (Barcelona)

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ABSTRACT

Urban agriculture is growing in cities and is rising to the roofs of buildings. The potential food contamination is a key issue to be resolved to guarantee the health of consumers, and it affects both urban agriculture promoters and consumers. Crop contamination from the soil can be overcome by adopting a soilless cultivation system that, with good management practices, can also avoid contamination from the fertirrigation system and pest treatments. It has recently increased the number of soilless cultivation systems in cities due to the good features it offers. This study focuses on the potential contamination of heavy metals in hydroponic lettuce crops due to atmospheric pollution in high-traffic areas. The contents of heavy metal in the air and the lettuce leaves were measured at 4 sites: a periurban-integrated rooftop greenhouse, a periurban rooftop, an urban courtyard and an urban rooftop. High-volume sensors were used to assess air contamination. Lettuce leaves were analysed to evaluate the heavy metal concentrations.

The results show that the heavy metal concentration in lettuce leaves is also below the EU-legislated limit in all studied cases. Specifically, the concentrations below the detectable analytic values were <0.02 mgNi/kg, <0.008 mgHg/kg, 0.005mgAs/kg and <0.005 mgCd/kg. The Pb concentration ranged from 0.0060 mg/kg to 0.0244 mg/kg. Although the chosen sampling locations were close to high-density roads and they are more vulnerable to a high concentration of metals, in the 4 sampling points heavy metal concentration in the air were less than 50% of the limits established in the legislation as the *lower assessment threshold.* This study concludes that the heavy metal content in the air of Barcelona is low and is not a source of contamination for urban crops including high traffic areas.

1. Introduction

1.1. Urban agriculture

Urban agriculture (UA) has been spreading throughout cities in the last few years. This is a response to the growing urban population, improved food safety and a reduction in poverty (UN-Habitat, 2013). At the same time, UA is presented as a tool for facing the increasing competition for resources associated with conventional agriculture (water, energy, soil) (Altieri et al., 1999; Cerón-Palma et al., 2012; Sanyé-Mengual et al., 2013; Specht et al., 2014). Furthermore, it increases the awareness of the environmental effects of the industrial food system and the need to address the social requirements of cities' inhabitants. (FAO, 2010).

Currently, different types of UA have been developed in urban areas. The large development of urban agriculture has led to the use

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of the roofs and courtyards of buildings in cities, where professionals and growers have found normally unoccupied open spaces for producing food (Weatherell et al., 2003). This is described in the literature as building-based agriculture (Sanyé-Mengual, 2015), and rooftop agriculture (RA) is the most common type (Thomaier et al., 2015). This subcategory includes all crop plants that are grown on the tops of buildings to take advantage of the available spaces on roofs. Among the different types of RA, there are rooftop greenhouses (RTGs). RTGs are built by installing greenhouses on the top of buildings to produce food with high yields using soilless growing systems (to reduce the structural load of the building), increasing the efficiency of resources and using constructed spaces that are currently unoccupied (Cerón-Palma et al., 2012). The first commercial RTGs were built in North America, and include Clendaniel (2011) in Brooklyn, USA, with an area of 1400 m^2, and Elton (2012) in Montreal, Canada with 2900 m^2 dedicated to the production of traditional crops (e.g., tomato, cucumber, pepper). These existing RTGs are constructed on roofs that are isolated from buildings. However, this kind of RTG can also be integrated (i-RTG) into a building, using the residual hot and cold air supplied by the building to provide an optimum range of temperatures for the crops and even using the greywater or rainwater collected in or on the building for irrigating the crops (Cerón-Palma et al., 2011; Nadal et al., 2017; Pons et al., 2015; Sanjuan-Delmás et al., 2018).

UA is an entire and unique ecosystem where the air-soil-plant system has a particular dynamic because of its anthropogenic sources. One of the aspects that needs to be improved is the safety of the food produced in the urban and periurban areas of cities, whether grown on land or in buildings. Furthermore, air pollution has been detected as an important factor in social perception studies of urban agriculture, where air pollution is perceived as having possible adverse effects on horticultural production and quality (Sanyé-Mengual et al., 2016; Specht et al., 2016). This leads to greater hesitance to implement it in different cities.

1.2. Air quality in European cities

According to (EEA, 2017), air pollution is a key environmental and social issue and is a complex problem that poses multiple challenges in terms of the management and mitigation of harmful pollutants. Air pollutants are emitted from anthropogenic and natural sources and are thus among the most important environment-related health concerns (European Environment Agency, 2008). The air pollution problems caused by toxic heavy metals such as As, Cd, Pb and Ni, in terms of air concentration, are much more localized than are other air contaminants. These emissions are closely related to particular industrial plants in the periurban area. Specifically, the concentrations of each metal in EU cities (619 city stations in 27 countries) in 2015 were as follows (EEA, 2017):

1. Arsenic: 94% of the stations reported a value below 2.4 ng/m^3 (lower than the assessment threshold defined in EU legislation (EU, 2004)). Seven stations reported concentrations above the target value (6 ng/m^3) in urban industrial areas in Belgium (3), Poland (2) and Finland (2).
2. Cadmium: 98% of the stations detected concentrations of Cd below 2 ng/m^3 (lower than the assessment threshold defined in EU legislation (EU, 2004)). Concentrations above the target value (5 ng/m^3) were measured at 6 stations, in suburban industrial areas in Belgium (3), Italy (1) and Spain (1).
3. Lead: 99% of the stations reported values below 250 ng/m^3 (lower than the assessment threshold defined in EU legislation (EU, 2004)). Levels above 0.5 μg/m^3 were detected only at a Belgian station.
4. Nickel: 97% of the stations reported concentrations of Ni below 10 ng/m^3 (lower than the assessment threshold defined in EU legislation (EU, 2004)). Concentrations of more than 20 ng/m^3 were detected at an industrial station in Norway and at another station in the United Kingdom.

Barcelona is one of the largest coastal European conurbations. Specifically, it is the densest Mediterranean coastal city, with approximately 1.6 million inhabitants in the central area and almost 5 million in the metropolitan area (Salvati et al., 2017).

The air quality in the city of Barcelona is representative of the EU cities. Specifically, the average values of each heavy metal during 2016 were as follows: 1 ng As/m^3; 0.4 ng Cd/m^3, 11.22 ng Pb/m^3 and 3.94 ng Ni/m^3 (Direcció General de Qualitat Ambiental, 2017). Moreover, Barcelona is a city where urban agriculture has developed extensively over the last several years. The Council of Barcelona has recently begun a plan to promote UA around the city. It has recognized and proposed as a future strategy working to advance/boost urban agriculture more than has been done to date. This will involve, among other steps, assessing the effects of environmental pollutants on crops and, consequently, on people's health ("Pla del verd i de la biodiversitat de Barcelona, 2020, 2013").

1.3. Heavy metal contamination effects on UA production

The origin of pollutants in the urban environment is largely human (Khan et al., 2008). Food contamination can occur either by contact with contaminated soils or by air pollution (Harrison and Chirgawi, 1989; Khan et al., 2016; Voutsa et al., 1996). Food contamination can be generated by different sources, such as wheeled transport emissions (Manta et al., 2002); previous land uses that were mainly industrial, atmospheric deposits from industrial activities and incinerators (Chen et al., 2005; Vittori Antisari et al., 2013); and management tasks such as fertirrigation (Khan et al., 2017) and pest treatments (Daddy Massaquoi, 2015; Peris et al., 2007).

Given the risk associated with their consumption on fresh vegetables, the European Union has set maximum lead and cadmium levels that can be found in vegetables (0.20 mg Pb/kg and 0.05 mg Cd/kg by weight in fresh vegetables) (EU, 2011). Obviously, these limits must also be observed in urban agriculture products. However, in many cases, UA is for direct personal consumption and not for sale. This means that there is no monitoring of these potential contaminants. In addition to Pb and Cd, potential health risks associated with other metals in edible vegetables have been detected (Khan et al., 2008; Vittori Antisari et al., 2015). Over the last several decades, different studies have focused on the contamination of urban agricultural products by analysing the end product for consumption (Table 1). In many cases, the soil is one of the most influential vectors in this contamination. When considering several influential variables (e.g., planting style, soil type), it is not possible to generalize the results to larger scales.

This paper focuses on the interaction between air quality and vegetable product quality to develop a new approach that will facilitate the planning of UA. As far as we know, there are no studies that have assessed these two issues together. This study uses a soilless cultivation system because we can remove the effects of soil as a contamination vector (Pennisi et al., 2016). Deepening the study of air pollution and the contamination of food simultaneously will allow the development of a map of the interaction between air quality and UA food in EU cities.

In UA, light substrates can be used to reduce the weight that the rooftops must bear (Thomaier et al., 2015). These commercial

Table 1
Summary of previous studies on UA food contamination. ☑ heavy metal evaluated in the literature.

Reference	Location	Study description	Heavy metal evaluated (1)	Crop contamination?	Source of contamination
(Sánchez-Camazano et al., 1994)	Spain (UE)	Analysis of soil and vegetables from 16 urban gardens.	☐ As ☐ Cu ☑ Pb ☑ Cd ☐ Hg ☐ Sn ☐ Cr ☐ Ni ☐ Zn	Cd	Traffic air contamination
(Kumar Sharma et al., 2007)	India	Soil crops in the periurban area of a high density city)	☐ As ☑ Cu ☑ Pb ☑ Cd ☐ Hg ☐ Sn ☐ Cr ☐ Ni ☑ Zn	Cd, Cu, Pb, Zn	Air contamination
(Säumel et al., 2012)	Germany (UE)	Comparison between UA food and market food	☐ As ☑ Cu ☑ Pb ☑ Cd ☐ Hg ☐ Sn ☑ Cr ☑ Ni ☑ Zn	For Pb in most cases	Not defined
(Vittori Antisari et al., 2015)	Italy (UE)	Effect of proximity to contamination sources (roads, railways) on tomato plants. Difference between soil and soilless crops.	☑ As ☑ Cu ☑ Pb ☑ Cd ☑ Hg ☑ Sn ☑ Cr ☑ Ni ☑ Zn	Just on soil crops near a road	Soil and air. Related to the site (proximity to a pollution source).
(Pennisi et al., 2016)	Italy (UE)	Analysis of the edible parts and roots. Cultivation in soil vs. peat in different systems of fertirrigation. Most contaminated allotment garden	☑ As ☑ Cu ☑ Pb ☑ Cd ☐ Hg ☐ Sn ☑ Cr ☑ Ni ☐ Zn	On soil crops; no on soilless crops	Soil
(Khan et al., 2017)	Pakistan	Uptake of hazardous elements by spring onion (Allium fistulosum L.) from soil irrigated with different types of water and possible health risk	☑ As ☑ Cu ☑ Pb ☐ Cd ☐ Hg ☐ Sn ☐Cr ☑ Ni ☐ Zn	Mo, Pb; The Ni, Zn, Cu and As sources were anthropogenic.	Mo, Pb fot Irrigation and anthropogenic

substrates are free of heavy metals, so this vector can be ruled out as a source of pollution for horticultural products. A vector that has been seen to have a large influence on contamination is the air (Sharmaa and Madhoolika Agrawala, 2007), so there is still a potential source of contamination (Voutsa et al., 1996). found that there is a high accumulation of Pb and Cd, especially in leafy vegetables, due to the atmosphere. Plants are interceptors of metal particulates deposited from the atmosphere. However, not all particulates intercepted are retained because some of them may be resuspended or washed away by rain (Harrison and Chirgawi, 1989). Various studies have shown that the concentration of heavy metals is inversely related to the distance from a road (Vittori Antisari et al., 2015; Gherardi et al., 2009; Naszradi et al., 2004). For example, Vittori Antisari et al., 2015, found that when vegetables are grown 10 m from the road, the risk of heavy metal accumulation is increased compared to that found 60 m from the road. It has also been determined that trees and buildings act as barriers to pollutants and markedly reduce traffic pollution (Säumel et al., 2012). Specifically, in the studied traffic areas, 50% of crops have Pb values that exceed the EU limits, in contrast to just 37% of the samples having critical Pb values when they were grown behind a barrier.

As mentioned earlier, few studies have analysed the relationship between the heavy metal concentration in the air and that in urban crops. Field surveys in urban areas are scarce but crucial for determining the health risk of UA. Nevertheless, as far as we know, the existing field studies did not sample air and food in the same place and at the same time.

This study was conducted in Barcelona, a city with air quality representative of most European cities. It focuses on the potential heavy metal contamination of UA crops by air pollution. The study simultaneously investigates urban and periurban areas with a high traffic density, so the potential for heavy metal contamination of food is high. To study the effect of air pollution on plants, an experimental design was used to remove other possible contamination vectors (e.g., soil, irrigation, pesticides). This new approach analyses air and food pollution simultaneously.

To address these gaps, this paper explores the following research objective:

- To determine the heavy metal concentrations in horticultural products grown on roofs in urban and periurban areas of Barcelona, with air pollution as the only source of potential heavy metal pollution.

Finally, considering the increase in RTG projects, it makes a first approach to study whether and how greenhouses can act as a barrier to air pollution of heavy metals in vegetables and the effectiveness of natural washing (rain) and manual washing at eliminating heavy metals.

2. Materials and methods

2.1. Research areas

To accomplish this study, 4 points were selected from urban and metropolitan areas of Barcelona. The points were chosen to evaluate if there are differences between the different locations of UA (urban vs periurban, rooftop vs courtyards and outside vs inside a greenhouse). The selected sites are all located near a high-density road (more than 50000 cars/work day). (Fig. 1).

The sites used for experimentation were as follows:

- A rooftop open air area in an ICTA-ICP building in the Universitat Autònoma de Barcelona (UAB) campus, 20 km from the Barcelona city centre, called the **Periurban-Rooftop site** (41.497681N, 2.108834E) This point is in the periurban area of Barcelona and is 240 m from an important highway (AP7-E90) and 20 m above street level. It has 14 lanes and a traffic density of 93127 cars/work days[1]
- An integrated rooftop greenhouse in the same ICTA-ICP building in the Universitat Autònoma de Barcelona (UAB) campus, called the **Periurban-i-RTG** (41.497530N, 2.108821E). This point is also 240 m from the AP7-E90 highway and is 16 m above the street level
- A particular courtyard within the centre of Barcelona, called the **Urban-Courtyard site** (41.38481N, 2.163125E). This point is near a main road of the city (Granvia Street) and is on the first floor above street level. Granvia Street has 8 lanes and a traffic density of 54210 cars/work day[2]
- A social orchard on the roof of the Municipal Institute for People with Disabilities (IMPD), called the **Urban-Rooftop** site (41.396970N, 2.169318E). This point is also near an important

street (Valencia Street) and is on the sixth floor above street level. Valencia Street has 4 lanes and a traffic density of 28267 cars/work day[2]

2.2. Experimental design

Every sampling point consisted of a crop area in which an air pollution sensor (high volume sampler) had been installed between the lines of crops. For the i-RTG, the air sensor was inside the greenhouse, such as the crops. For the last several years, in all areas, several soilless horticultural crops have been developed, guaranteeing the good development of plants.

The EU air legislation define the limits for the following metals: Ni, As, Cd and Pb (EU, 2004). The same heavy metals were considered for the air quality in this study. Despite EU legislation defining the limits for maximum levels of only Pb (0.1 mg/kg) and Cd (0.050 mg/kg) (EU, 2011) in foodstuffs, in this study, As, Ni and Hg were also studied.

To study only metal contamination from the air, all crops were planted in the same medium (sacks of perlite substrate, each with four units of lettuce). Perlite is an inert material that allows horticultural production without soil. This means that possible heavy metal contamination of plants via the roots and the soil is ruled out. Water and nutrition solutions for plants were provided via a drip fertirrigation system. In all cases, the nutrition solution was the same, and the irrigation doses were adapted to the needs of the crop (Sanjuan-Delmás et al., 2018).

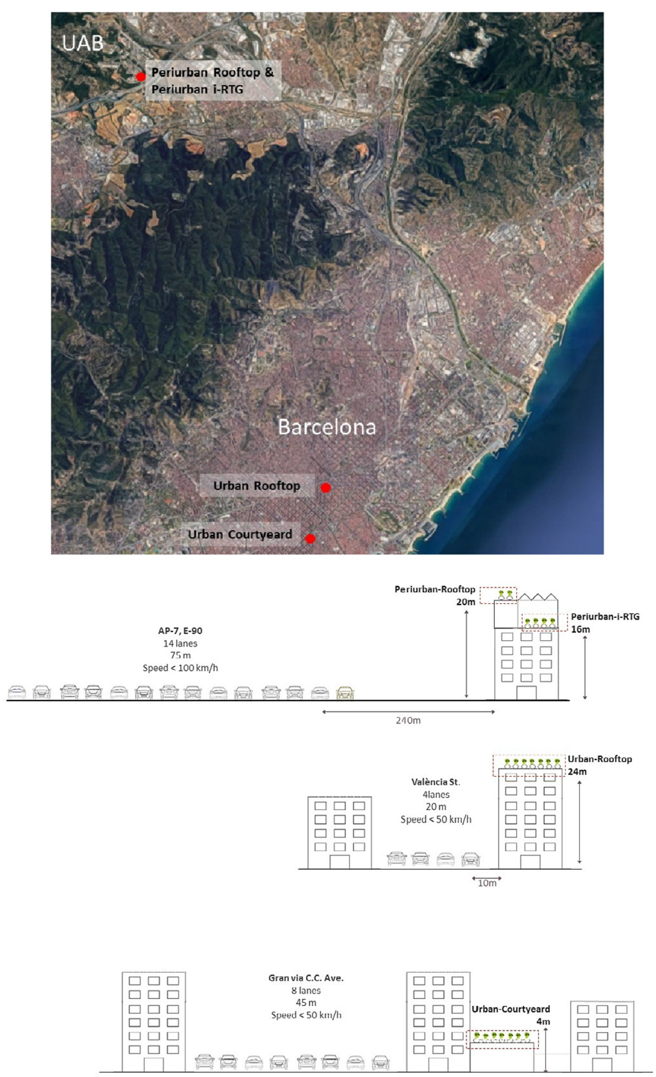

Fig. 1. Research areas and sampling points. Urban and periurban sites used for the experiment.

Oakleaf lettuces (*Lactuca sativa* var. *capitata L.*)) were planted simultaneously in the 4 locations. The crops were monitored and controlled following conventional agronomic guidelines for lettuce production. No chemical treatments were performed to avoid possible influences.

This study tests a new experimental installation and allows the simultaneous analysis of the air and the crops. Both the crops and the air were sampled (Fig. 2 and Table 2). Additionally, both the substrate and the water from the fertirrigation were analysed to ensure that they were not a source of heavy metal contamination.

The study was performed during two tests of lettuce crops. The first test was from 20/04/2017 to 29/05/2017 (40 days), and the second one was from 22/06/2017 to 25/07/2017 (34 days). The duration of the study was determined by the speed of growth of the crops. The tests ended when the crop reached commercial size. The lettuces were harvested at all locations and the air sensor systems were turned off on the same day.

2.3. Meteorological and climatic conditions

This study was carried out from April 2017 to July 2017. The summer season (June–August) is usually dry and is associated with high temperatures and low rainfall. Specifically, in Barcelona during 2017, an absolute maximum of 35.9 °C and an absolute minimum of 2.8 °C were recorded, with the annual average at 18.1 °C. Specifically, in the study period and in the two sampling areas, the following meteorological conditions were observed. In the city of Barcelona (Raval station) the average temperature was 22.18 °C, the maximum was 33.3 °C and the minimum was 7.9 °C. The total precipitation during this period was 73.3 mm (Ruralcat, 2017). In the metropolitan area, near the building where the study was carried out, the average temperature was 20.49 °C, and the minimum and maximum were 0.7 °C and 35.9 °C, respectively. The accumulated rainfall during the period was 93.1 mm.

During the study period, the number of rainy days and the amount of rainfall were tracked to assess the effect rainwater on crop contamination. In this case, the most important effect of rainwater was the washing effect in the leaves. Rain water can also transport solution elements that can function both as nutrients and as contaminants. Nevertheless, the amount of water that could reach the substrate was very small because crops were cultivated on closed perlite bags.

2.4. Sample collection

2.4.1. Crop sampling

The crop samples were taken at random from different rows in the growing areas, ignoring the edges of the planting. To make each sample more representative, each lettuce sample consisted of five lettuce plants picked at random from the same row of the crop when they reached commercial size (>200 g).

During the first test, the effects of washings on the removal of heavy metals from the vegetable surface were assessed. In this case, double samples were collected at each point and then divided into two groups. The first group was kept as they were before being chopped into small pieces and mixed, whereas the edible portions of the lettuce in the other group, after harvesting and transportation to the laboratory, were prepared for analysis and rinsed

Fig. 2. Example of the experimental design. Left: Image from the periurban rooftop during the first test. Middle: high volume sensor. Right: glass microfibre filters after 48 h of sampling.

Table 2
Characteristics of crop samples and air samples in each conducted test: Test 1: 20/04/2017 to 29/05/2017 and Test 2: 22/06/2017 to 25/07/2017. ☑ Analysis conducted.

Points of sampling		Samples	Lettuce washed leaves
Periurban-Rooftop	Test 1	☑ Lettuce	☑ Washed
		☑ Filter Air	☑ Unwashed
	Test 2	☑ Lettuce	☐ Washed
		☑ Filter Air	☑ Unwashed
Periurban-i-RTG	Test 1	☑ Lettuce	☑ Washed
		☑ Filter Air	☑ Unwashed
	Test 2	☑ Lettuce	☐ Washed
		☑ Filter Air	☑ Unwashed
Urban-Courtyard	Test 1	☑ Lettuce	☑ Washed
		☐ Filter Air	☑ Unwashed
	Test 2	☑ Lettuce	☐ Washed
		☑ Filter Air	☑ Unwashed
Urban-Rooftop	Test 2	☑ Lettuce	☐ Washed
		☑ Filter Air	☑ Unwashed

with tap water to remove surface dust, as is normally done in the kitchen. Then, the latter were also chopped into small pieces and mixed. In the second test, the suitability of repeating the experiment was reconsidered, taking into account the effect of manual washing in the first test.

2.4.2. Air sampling

To collect air samples, high-volume sensors (MCV CAV-A/mb, ©MCV) were used, working at a volume of 30 m^3/h in 48-h periods, using glass microfibre filters (150 mm Ø(UNE-EN 12341:2015; UNE-EN 14907:2006a,b). Every 48 h, the filter was removed manually and changed for a new one. Once all the filters were collected throughout the period, they were crushed, homogenized and analysed. The analysed air samples are the average value for the whole period of the crop.

2.4.3. Substrate and water sampling

Three substrate samples (*Substrate-bag1, Substrate-bag2* and *Substrate-bag3*) were collected from 3 randomly selected bags and analysed following the methodology described in (Sanjuan-Delmás et al., 2018) to ensure that there was no contamination from the substrate to the crop. Each bag was opened and spread, mixing the perlite and making a layer with uniform thickness. For each sample, the roots of the plant were carefully removed. To take a representative sample, this layer of perlite was divided into 20 sections, and approximately the same amount was taken from each section with a spoon. The sample was then carried to the lab in a plastic jar, and a similar process was conducted using a plastic tray to obtain a smaller sample for analysis. The sample was then dried at 383 K and ground using an analytical mill.

The building contains a rainwater harvesting system that collects the rainwater from the roof of the building. This untreated rainwater was used in the i-RTG to irrigate the crops after addition of mineral fertilisers. Three water samples (Irrigation-water1, Irrigation-water2 and Irrigation-water3) were collected from the irrigation drippers and analised. Thereby, the heavy metal contribution of the irrigation water (rain water harvested plus mineral fertilisers) was evaluated.

2.5. Chemical analysis

Air and crop sample analyses were carried out by the Agency of Public Health of Barcelona with ENAC accreditation (UNE-EN ISO/IEC 17025:2005).

Air Sampling: The following metals were analysed: Ni, As, Cd and Pb. A microwave digestion of the samples (0.2 g of air sample) was performed with 10 mL of 20% suprapur nitric acid at up to 200 °C (15 min to arrive to 200 °C and 15 min at 200 °C). Water was added to the digested extracts to reach a final volume of 40 mL (instrument Milestone Ethos One). The elements were determined by inductively coupled plasma mass spectrometry (ICP-MS), using helium in the collision cell to eliminate poliatomic interference (ICP-MS Agilent 7700). The quantification was done with an internal standard (103Rh). The masses used for each analyte were 60Ni, 75As, 111Cd, 208 Pb, and 201Hg.

To compare the results with the usual heavy metal concentrations in the areas under study, the mean concentration of each studied metal from 01-01-2016 to 31-03-2017 was measured in Barcelona and Castellbisbal (the closest station to the periurban area under study).

To rule out any contamination during the process of sampling and the treatment of the samples, the concentration of heavy metals was analysed in four field blanks. Since the filters used were fiberglass filters, they contained a concentration of cadmium (5.7 μg Cd/g filter). A contamination of Ni was also produced during the sampling process. On average, the values detected in the blanks were 9 μg Ni/g filter. These values were subtracted from the concentrations detected in the study samples.

Crop Sampling: The following metals have been analysed: Ni, Hg, As, Cd and Pb. Also a microwave digestion of the samples (0.50 g of vegetable sample) was done with 10 mL of 20% suprapur nitric acid at up to 200 °C (15 min to arrive to 200 °C and 15 min at 200 °C). Water was added to the digested extracts to reach a final volume of 40 mL (instrument Milestone Ethos One).

Instrumental determination was done by ICP-MS (ICP-MS Agilent 7700) using helium in the collision cell to eliminate polyatomic interference. The quantification was done with an internal standard. For food samples, the internal standards were 72Ge for Ni and As, 103Rh for Cd and 209Bi for Pb and Hg.

Substrate and water sampling: Water samples were collected, filtered, and stored at 4 °C until analysis in 100 mL polypropylene pots. The total heavy metals in irrigation water were analysed by ICP-AES.

The samples of perlite were digested in duplicate with concentrated HNO_3 in a microwave oven together with digestion blanks. Then, they were lixiviated and the solids were removed, leaving a liquid solution with the extracted nutrients from the perlite, which was then analysed using ICP-AES.

2.6. Data processing and statistics

Crop analyses were carried out in triplicate and were expressed using average values and standard deviations. When heavy metals were detectable by the analytic equipment, the R Studio software was used to determine significant differences between the sampling points and between the unwashed and washed plant samples. This was tested using a one-way analysis of variance (ANOVA) (Tukey's test, $p < 0.05$). Before statistical analysis, the assumptions of ANOVA were checked by a Shapiro-Wilk test.

3. Results and discussion

3.1. Heavy metal concentrations in the irrigation water and substrate

This study rules out the medium having any influence on the concentration of heavy metals in the crop. In this case, the substrate and the irrigation water can be disregarded as contamination vectors. Thus, the only vector that could cause contamination is the air. Table 3 shows the results of the heavy metal concentrations in the irrigation water and the perlite bags. The substrates no exceeded the limit value established by legislation (RD 865/2010, 2010). Moreover, the levels of all heavy metals are under the limits of detection (Table 3). The irrigation water is used to compare the limits established in BOE (BOE, 2007). Specifically, limits are defined for irrigating crops with a water application system that allows direct contact between the regenerated water and the edible parts for fresh human consumption. All samples no exceeded these limits.

3.2. Air quality

Table 4 shows the air quality results obtained at the sampling points and in the different tests. The values recorded in each context are shown, as are the limits established by the EU legislation (EU, 2004) for each of the pollutants studied (Ni, As, Cd and Pb). None of the metal concentrations in the air are above the target value. 'Target value' means a concentration in the ambient air set with the aim of avoiding, preventing or reducing harmful effects on human health and the environment as a whole, to be attained where possible over a given period (EU, 2004). Although the places are all close to the road and are thus more likely to have high concentrations of metals (Vittori Antisari et al., 2015), this is not the case. In particular, in all locations, the heavy metal concentration is lower than 50% of the limit established in the legislation as the lower assessment threshold, which is defined as a level specified in Annex II of the Directive below which the sole use of modelling or objective estimation techniques shall be possible to assess ambient air quality, in accordance with Article 6 (4) of Directive 96/62/EC. This is the most restrictive value of the legislation.

The values of heavy metals detected in the air are similar to or slightly higher (in the case of Cd) than the average values observed in Barcelona and its surrounding areas in recent years (0.39 ng Cd/m^3 annual average 2017). As mentioned above, this may have occurred because places close to high traffic roads were chosen for this study. As shown in the introduction, the air quality in Barcelona is similar to that of most European cities. Therefore, the results of the interaction between air and urban agriculture can serve as a reference for other cities.

It is also important to note that the concentrations of heavy metals at all sampling points are very similar, including for the areas studied and for the air inside the greenhouse.

In fact, the concentrations of Pb and Cd inside the greenhouse are slightly higher (9% and 19%, respectively, in the first test) and those of As and Ni are slightly lower (9% and 36%, respectively, in the first test) than are the concentrations outside the greenhouse. According to these results, there is no barrier effect on the air quality when installing a greenhouse for rooftop agriculture. It should be highlighted that it is a greenhouse with higher ventilation rates than a conventional greenhouse (Montero et al., 2017; Nadal et al., 2017) Specifically, during the studied period, the greenhouse was semi-opened for 23% of the time and completely opened for 74% of the time. This implies more air renewal, which can result in no differences in indoor and external air quality. However, a previous study on the aerobiological air quality carried out in the same greenhouse (Ercilla-Montserrat et al., 2017) found that there were differences between the interior and external air of the greenhouse. This divergence can be due to two reasons. The first is that the concentration of heavy metals in the air is very low, which makes it difficult to detect differences between the sampling points. It is also possible that the dynamics of the two kinds of contaminants (heavy metals and biological particles) make the difference, considering their weight, adherence to other solid particles, and other factors.

3.3. Heavy metal content in lettuce tissues

The metal concentrations of Ni, Hg, As and Cd were not detected

Table 3
Heavy metal concentrations in irrigation water (μg/l) and in the perlite bag (mg/kg sms). 1. Heavy metal limits in regenerated water for crop irrigation (BOE, 2007). Spain limit for class A substrates (RD 865/2010, 2010).

	Ni μg/l	Hg μg/l	As μg/l	Cd μg/l	Pb μg/l
Irrigation-water1	<5.00	<1.00	<1.00	<5.00	<10.0
Irrigation-water2	5.70	<1.00	<1.00	<5.00	<10.0
Irrigation-water3	<5.00	<1.00	<1.00	<5.00	<10.0
Spain limit value (regenerated water)	200	–	100	10	–
	Ni mg/kg	**Hg mg/kg**	**As mg/kg**	**Cd mg/kg**	**Pb mg/kg**
Substrate-bag1	<5	<0.4		<0.50	<5
Substrate-bag2	<5	<0.4		<0.50	<5
Substrate-bag3	<5	<0.4		<0.50	<5
Spain limit value[2]	25	0.4		0.7	45

Table 4
Summary of target values and assessment thresholds in EU legislation and heavy metal concentrations in the sampled air. Legend: 1. Data from EU (EU, 2004) 2. Annual average (2016) of concentrations of heavy metals in the 10 city measurement stations; data are derived from Xarxa de Vigilància i Previsió de la Qualitat de l'Aire (XVPCA) 2017.

		Ni (ng/m3)	As (ng/m3)	Cd (ng/m3)	Pb (ng/m3)
EU legislation	Target value UE[1]	20	6	5	500
	Upper assessment threshold UE[1]	14	3,6	3	350
	Lower assessment threshold UE[1]	10	2,4	2	250
Periurban Rooftop	Test 1	2,51	0,83	0,71	10,55
	Test 2	4,92	0,67	0,29	7,52
Periurban i-RTG	Test 1	0,75	0,76	0,77	11,13
	Test 2	1,26	0,69	0,47	6,70
Urban Courtyard	Test 2	4,53	0,68	0,28	8,70
Urban Rooftop	Test 2	3,32	0,71	0,39	7.10
Barcelona average[2]		3,74	1,00	0,39	10,75
Maximum Barcelona		6,89	1,38	0,64	26,58
Minimum Barcelona		2,27	0,72	0,15	6,23

in any analysis of the lettuce. Thus, these heavy metals may not have significant accumulation levels. This type of crop was selected because leaf crops generally have more metal (Cd, Co, Cr, Cu, Fe, Mn, Ni and Pb) absorption and/or accumulation than inflorescence in their edible parts. The high heavy metal contents in leaf crops seem to be due to metabolic processes, as these crops are accumulators. Furthermore, these crops can easily absorb heavy metals that are deposited on their leaves by atmospheric deposition (Peris et al., 2007). At the same time, as lettuce is a fast-growing crop, it is possible to detect a rapid response to the effect of heavy metals without other types of interference, as is possible in crops with slower development.

In the case of Cd, the concentration values are at least one order of magnitude lower than the target value defined by EU legislation (0.05 mg Cd/kg). The Hg and As limits are not defined in the EU legislation. The Pb concentration ranged from 0.0060 mg/kg to 0.0244 mg/kg. In all cases, the Pb concentration was below the EU limit (0.1 mg/kg). Specifically, the Pb concentration detected was <25% of the legal limit (EU, 2011).

Both cadmium and nickel are elements that have a high rate of penetration in leaves. In other words, they are absorbed by the plant once they reach the leaves (Pennisi et al., 2016; Voutsa et al., 1996). The concentrations of Cd and Ni in the air were under the legal limits (Table 4), and the concentrations detected in the crops were practically zero. Even in the case of Ni, which was found in the air in a greater concentration than in the average of Barcelona, the concentration was under the detected value in the crop samples.

In general, the results demonstrate that in Barcelona, UA production can be developed with confidence if care is taken to work with substrates, irrigation water, nutrients and pesticides that are free of metallic contaminants. This means that actual air pollution does not contribute to crop contamination. These results concur with those of (Pennisi et al., 2016), who found that crops grown in a soilless environment have metal contents of −70% for Cr, −61% for Cu, −45% for Cd and −81% for Ni, which are lower than the contents of crops those grown in soil (Vittori Antisari et al., 2015). detected a high concentration of heavy metal in crops grown near a road in Bologna (up to 8 mg/kg). This finding concurs with previous literature and evidence that the major contamination risk in urban areas is related to soil pollution (Peris et al., 2007). and (Finster et al., 2003) found that soil is the source that is most responsible for crop contamination.

Using a soilless system, soil contamination can be avoided; in this case, the air is the most important potential source of heavy metal in vegetables. For this reason, our heavy metal concentration results are much lower than the results from other studies in European cities. Other studies have also detected that agronomic practices and atmospheric deposition may influence crop accumulation. Specifically (Peris et al., 2007), studied the heavy metal content of horticultural crops in European Mediterranean region. In this study, it was detected that the concentration exceeds the legal values in lettuce and Swiss chard grown in an area influenced by industry and/or roads; the mean contents in lettuce were 0.21 ± 0.31 mg Cd/kg and 0.29 ± 0.27 mg Pb/kg.

Although this study was conducted in potentially unfavourable air conditions, the absence of any heavy metal contamination in lettuce is very positive. Despite having chosen points differentially susceptible to contaminated air, the concentrations of most of the heavy metals studied were below the detectable values, and although Pb was the most common contaminant in all samples, its level was below 25% of the legal value. This occurred because the heavy metal concentrations in the air are much too low to have an influence on vegetable contamination. They were lower than the EU target values and similar or slightly higher than the average values observed in Barcelona and its surrounding areas (see Annex I). Finally, we must highlight that in recent years, the air quality in

Barcelona and the whole metropolitan area has generally been below the legal threshold (Direcció General de Qualitat Ambiental, 2017).

Lead is the only heavy metal detected in the lettuce leaves, but in all cases, it was below the threshold established by legislation (the values oscillate between 6% and 24% of the limit value defined by the EU) (Table 5). As demonstrated previously, little of this element was detected in the air during this study, and the values detected were similar to the average values for the city of Barcelona and other EU cities. Pb is the metal that had the greatest presence, and this concurs with (Harrison and W.R.J, 1987), who demonstrated that the atmospheric contribution of Pb in the exposed parts of plants was between 45 and 97% of the total metal content. This results concord with the conclusions of (Voutsa et al., 1996) because in his study, the highest accumulation of Pb in lettuce was due to atmospheric deposition. In agreement with these previous studies, the present results suggest that if the air had a higher Pb concentration, the values detected in the leaf would also be higher. The results of this study agree with the results of recent studies, which showed that the concentration of lead in crops decreased. This is partly due to new air quality legislation that has changed the fuel used in gasoline cars. Prior to 2001, small amounts of lead were added to gasoline to improve the performance of the engines. However, as a polluting metal, the European Union, through Directive 98/70/CE, banned the sale of leaded gasoline on January 1, 2000. In Spain, this ban came into effect in August 2001 (Mielke et al., 2011). found that the lead concentration in plant tissue has been reduced by the adoption of unleaded gasoline. Therefore, in studies conducted before this law, the concentrations of lead were higher. For example, in 1994 (Sánchez-Camazano et al., 1994), detected that the Pb concentration in vegetables from urban gardens was 2.60 times higher than that in natural soils because urban pollution arises mainly from the diffusion of Pb during gasoline burning. The Pb contents of vegetables ranged between 4.17 and 52.7 mg/kg; these values greatly exceeded the actual legal limit. To determine how much influence the soil properties have on the crop contamination, its correlation with soil contamination was explored, and no significant correlation was found. This was possibly because Pb is a deposition pollutant, as detected in this study. This result concur with those of (Alegría et al., 1991), who detected a correlation between soil contamination and vegetable content for Ni and Cd but not for Pb. Anthropogenic and environmental factors were the most important sources for the Pb content of vegetables.

According to these results and because Barcelona air quality is representative of EU cities (EEA, 2017), it is possible to make a first approximation and affirm that RA developed in the EU will not accumulate heavy metals due to the air quality, if the other possible sources of contamination are controlled.

3.4. Washing effect

Due to the low levels of heavy metal absorption (below the level of detection of the analysis equipment), the effects of washing the lettuce cannot be observed for most of the heavy metals studied. Nevertheless, as in previous studies, an influence of the washing effect was detected, so it was considered important to account for this aspect. Only for Pb can an initial conclusion be drawn. Pb has been found to be deposited on leaves with dust if the plant is not washed either by rain (Harrison and Chirgawi, 1989; Voutsa et al., 1996) or manually (Finster et al., 2003; Kim et al., 2015). This means that an unwashed lettuce plant contains greater quantities of Pb. Table 6 shows the results and presents the differences between washed and unwashed lettuce.

3.4.1. Manual washing

The results of this experiment show that there is no statistically significant difference (p value < 0.05) between unwashed lettuce and manually washed lettuce in the three sampling points studied during the first test.

This finding implies that there are no differences detected due to this process. This outcome, however, contrasts the results of (Kim et al., 2015), who detected higher concentrations of heavy metals in unwashed lettuce samples than in washed samples in Seoul. Specifically, in that study, compared to unwashed plants, washing reduced the hazard quotient of heavy metals by 17–28%. However, it is important to highlight that the values reported in the aforementioned study ranged from 7.94 to 13.76 mg Ni/kg-1 dry weight for nickel and from 1.41 to 2.6 mg Pb/kg^{-1} dry weight for lead. This was due to the city's high volume of traffic and various types of industrial activities and the concentration obtained in Kim's study was 20–35 times higher than that in our study for Ni and for Pb, it was 3–5 times higher.

Potential effect of greenhouse infrastructure as a barrier.

This is the first study to assess the effect of the greenhouse infrastructure as a barrier to heavy metals. Before the experiment was conducted, there were two unknown issues to solve.

1. Is there a difference in heavy metal concentrations between the air inside the greenhouse and the outside air? In other words, does the greenhouse infrastructure prevent the penetration of heavy metals by air?
2. Does rainwater act as a source of metal deposition into the lettuce or as a cleaning agent?

The first question has been answered in section 3.2. *Air quality.*

Table 5
Heavy metal concentrations (Ni, Hg, As, Cd and Pb) in lettuce samples in the urban and periurban sites under study and the EU-defined limits in leaf crops (EU, 2011). U: unwashed; W: washed.

		Ni (mg Ni/kg sample)	Hg (mg Hg/kg sample)	As (mg As/kg sample)	Cd (mg Cd/kg sample)	Pb	
						(mg Pb/kg sample)	% from EU legislation
Periurban Rooftop	Test1 U	<0.020	<0.008	<0.005	<0.005	0.0090	9%
	Test1 W	<0.020	<0.008	<0.005	<0.005	0.0080	8%
	Test2 U	<0.020	<0.008	<0.005	<0.005	0.0228	23%
Periurban i-RTG	Test1 U	<0.020	<0.008	<0.005	<0.005	0.0060	6%
	Test1 W	<0.020	<0.008	<0.005	<0.005	0.0070	7%
	Test2 U	<0.020	<0.008	<0.005	<0.005	0.0090	9%
Urban Courtyard	Test1 U	<0.020	<0.008	<0.005	<0.005	0.0110	11%
	Test1 W	<0.020	<0.008	<0.005	<0.005	0.0090	9%
	Test2 U	<0.020	<0.008	<0.005	<0.005	0.0244	24%
Urban Rooftop	Test 2 U	<0.020	<0.008	<0.005	<0.005	0.0187	19%
EU legislation		–	–	–	0.050	0.1	

Table 6
Rainfall parameters and Pb concentration in the air and in the crop. *Days where the rainfall exceeded 3 mm are in parentheses *U: unwashed; W: washed.*

		Lettuce sample (mg Pb/kg)	Air sample (ng Pb/m3)	Raining days*	Accumulated rainfall (mm)
Periurban Rooftop Open air	Test1 U	0.009	24.41	10 (3)	39.4
	Test1 W	0.008			
	Test2 U	0.023	21.18	5 (2)	26.4
Periurban i-RTG	Test1 U	0.006	26.69	No rain (inside greenhouse)	No rain (inside greenhouse)
	Test1 W	0.007			
	Test2 U	0.009	20.66		
Urban Courtyard	Test1 U	0.011	–	7 (3)	38.6
	Test1 W	0.009	–		
	Test2 U	0.024	22.44	6 (3)	22.8
Urban Rooftop	Test2 U	0.019	20.76	6 (3)	22.8

According to these results, there is no barrier effect due to the greenhouse.

Regarding the second question, as was also found by washing plants manually, it is not possible to determine if there is a barrier effect due to the greenhouse in the studied metals, except in the case of Pb. This is because the concentrations of Ni, Hg, As and Cd were below the level of detection.

Table 6 lists the total Pb contents of the sampling points. The results show that the differences detected between the Pb concentrations on the lettuce can be related to the washing effect of rainwater and the deposition of Pb by the dust. In the first test, the lettuce grown inside the greenhouse had the same Pb concentration as did those grown outside. Nevertheless, during the second test, the situation changed: the Pb concentration in lettuce grown inside the i-RTG was lower than that in lettuce from outside. If the two tests are compared, it is observed that although the concentration of Pb in the air was similar during the two tests, the concentrations detected in the vegetables in the outdoor experiments were much higher in the second test. Specifically, for the periurban rooftop site, the concentration in vegetables was 2.67 times higher in the second test than in the first one. For the urban courtyard site, the concentration was 2.44 times higher in the second test. A first interpretation relates the air quality, heavy metal concentrations in crops and rainfall patterns. In Barcelona, there is a marked predominance of anticyclonic situations, with stable weather that results in many days without notable atmosphere movements. Therefore, there are long periods of stagnation and a lack of air mass exchanges. This situation is reinforced by the influence of the orography of the metropolitan area of Barcelona. A direct effect of this atmospheric stagnation is the accumulation of particles derived from urban metabolism. As a consequence, air quality decreases progressively until the atmospheric dynamics change (mainly with episodes of instability and rainfall) to favour the dispersion of pollutants and the renewal of air masses (Capdevila et al., 2017). In this study, during the first test, there were 10 days of rain and a total rainfall of 39.4 mm, mostly concentrated in 4 days. During the second one, there were 5 days of rain and a total rainfall of 26.4 mm, mostly concentrated in 2 days. Thus, there were important differences in air renewal.

More research is needed to confirm these results, including measuring all heavy metals in different meteorological conditions with different rainfall patterns and more extreme anticyclonic conditions.

4. Conclusion

This study has proven the feasibility of growing leaf crops on the rooftops of Barcelona and its surroundings using soilless systems in high-traffic areas. Previous studies have suggested that the air pollution and, specifically, the heavy metal concentration in city air are perceived to have a possible adverse effect on horticultural production and quality (Sanyé-Mengual et al., 2016; Specht et al., 2016). The approach proposed in this study consists of simultaneously analysing air quality and food quality in the same places and at the same time in urban areas. Soilless systems (as an alternative to soil agriculture) can avoid introducing heavy metals into crops because it is possible to guarantee non-contaminated substrates and other system inputs.

This research provides a new approach that can be integrated into UA development policies, considering the heavy metal levels in air pollution for urban planning with hydroponic crops.

In general, the range of trace metal contents in edible parts of the analysed hydroponic crops (lettuce) is lower than the concentrations reported in previous studies (Pennisi et al., 2016; Säumel et al., 2012; Sharmaa and Madhoolika Agrawala, 2007; Vittori Antisari et al., 2015) because most previous studies were conducted on city soils or in beds filled with commercial soils. In both cases, the soils or the substrates contain high heavy metal concentrations. In this study, the heavy metal concentrations in lettuce is below the EU-legislated limits in all studied cases. Specifically, the heavy metal accumulation (Ni, Cd and As) in lettuce grown in rooftop agriculture in open-air and inside i-RTGs in Barcelona and its periurban area is under the detectable levels (<0.02 mg Ni/kg, <0.008 mg Hg/kg, 0.005 mg As/kg and <0.005 mg Cd/kg that is a 10% of the UE limit). Pb has the highest concentration, which ranges from 0.0060 mg/kg to 0.0244 mg/kg (between 6% and 24% of the EU limit); thus, this heavy metal also does not have a significant accumulation level.

The air quality in Barcelona and its surroundings is representative of EU cities (Direcció General de Qualitat Ambiental, 2017; EEA, 2017). The Ni, As, Cd and Pb concentrations detected in the air are less than 50% of the limits established in the legislation as the *lower assessment threshold* (EU, 2004). Although the chosen sampling locations were close to high-density roads, they are more vulnerable to a high concentration of metals (Vittori Antisari et al., 2015).

We further confirm that the air quality in Barcelona and its surroundings is not a limiting factor for the development of UA, even though the sampling points were close to high-density roads. Thus, the results of the possible contamination of crops due to air pollution can be applicable to other urban areas of the EU.

This new approach, which considers the contamination of the air and the crop simultaneously, is very useful for promoting urban agricultural development plans. This is especially interesting in cities with high concentrations of heavy metals in the air. This new proposal can be replicated with other pollutants such as AOT40, SO_2, NO_2, and acid rain. The possibility of generating new maps of distribution of UA taking into account the air quality will contribute to giving new decision-making tools for individuals and administrators.

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