https://doi.org/10.5194/egusphere-2022-174
Preprint. Discussion started: 19 July 2022

Seasonal variation of mercury concentration of ancient olive groves of Lebanon.

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https://doi.org/10.5194/egusphere-2022-174
Preprint. Discussion started: 19 July 2022

Abstract. This study aimed to investigate the olive, an iconic tree of the Mediterranean basin, seasonality of (Hg) mercury pollution. Hg concentrations of foliage, stems, soil surface, and litter was analyzed on monthly basis in ancient olive trees growing in two groves in Lebanon, Bchaaleh and Kawkaba (1300 and 672 m.a.s.l respectively). A significantly lower concentration was registered in stems (~7-9 ng/g) with respect to foliage (~35-48 ng/g) in both sites with the highest foliage Hg concentration in late winter-early spring and the lowest in summer. It is noteworthy that olive fruits also have the lowest Hg concentration (~7-11 ng/g). The soil has the highest Hg content (~62-129 ng/g) likely inherited through the cumulated litter biomass (~ 63-76 ng/g). A good covariation observed between our foliage Hg time-series analysis and those of pCO_2 and Hg concentrations of the atmospheric Northern hemisphere confirms that mercury pollution can be studied through olive trees. More precisely, spring sampling is recommended if the objective is to assess the tree's susceptibility to Hg uptake. This may draw an adequate baseline for global inventories on Hg vegetation uptake and new studies on olive trees in the Mediterranean to reconstruct regional Hg pollution concentrations in the past and present.

Keywords: Eastern Mediterranean, ancient groves, *Olea europaea* L., mercury pollution, plant tissues, soil and litter

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1. Introduction

Mercury (Hg) is among the most common heavy metals polluting the Earth (Briffa et al. 2020). It is found as all heavy metals naturally on the Earth's crust reservoir and in the atmosphere through the natural long-term Hg biogeochemical cycle (i.e., volcanic activities, geological weathering). This metal is easily modified into several oxidation states and it can also be spread through many ecosystems (Boening 2000). The natural Hg cycle has been modified due to anthropogenic activities (i.e., mining, smelting, soil erosion due to deforestation, gold extraction, agriculture-fertilizers, manure) (Patra and Sharma 2000). Among natural and anthropogenic Hg emissions, inorganic elemental Hg (Hg(0)) is the most dominant chemical form. It is generally transferred through the atmosphere by the winds and mass air. This highly diffusive Hg can easily pass biological barriers (i.e. cell membranes, foliage, skin) and bind covalently with organic groups forming the widespread toxic methylmercury (MeHg, CH_3Hg^+) (Clarkson and Magos 2006). Anthropogenic activities endorse an accumulation of heavy metals in terrestrial and aquatic ecosystems. This can remain for over 100 years after the closure of the pollution source. The exchange between the Hg amount in the soil and uptake of the Hg by the plants is not stable and is variable dependent (e.g. cation-exchange capacity, soil pH, soil aeration, and plant species) (Patra and Sharma 2000).

Large forests with large foliage surface area are known to act as a sink of atmospheric Hg in the ecosystem. Plant foliage take up Hg deposited on leaf surfaces through the stomata (ie. Photosynthesis) and leaf cuticles (Hanson et al. 1995; Jiskra et al. 2018; Li et al. 2017; Lodenius et al. 2003; Maillard et al. 2016; Rea et al. 2002; Yanai et al. 2020) where it accumulates with minimal mobility and small portions released back into the atmosphere or transfers the Hg through the other plant organs (Cavallini et al. 1999; Hanson et al. 1995; Li et al. 2017; Lodenius et al. 2003; Schwesig and Krebs 2003). Water and soil are another uptake Hg source through roots (Bishop et al. 1998; Li et al. 2017). These two Hg absorption pathways in plants are also described in Hg contaminated sites on several plant species reporting the accumulation of Hg in the foliage and also in the roots where Hg can be absorbed through xylem sap (Assad et al. 2017). Tree's Hg is transferred to the forest floor through litter and throughfall and hence passes to the soil (Rea et al. 1996). Litter is said to constitute 30 to 60 % of the Hg atmospheric deposition in Europe and North America forests (Rea et al. 1996; Blackwell and Driscoll 2015; Zhou et al. 2018). The Hg input through the litter is greater than the input from that of the wet deposition (Wang et al., 2016). The Litter Hg is the dominant pathway in forests where it contributes to 53 and 90 % of the dry deposition to the forest (Wright et al., 2016).

In earth ecosystem, soil as part of the geological reservoir has naturally the highest Hg reservoir (D. Obrist et al., 2018; O'Connor et al., 2019) followed by trees (Yang et al. 2018). This Hg is provided by natural geological sources and natural events such as forest fires, volcanic eruptions (Ermolin et al. 2018; Obrist et al. 2018; O′Connor et al. 2019) and anthropogenic sources (UNEP, 2019). In return, soil and plants can reemit Hg to the atmosphere through diffusion and transpiration during photo-respiration biological processes (Luo et al., 2016; Yang et al., 2018a; Assad, 2017; Schneider et al., 2019; Gworek et al., 2020). Trees are hence important drivers of Hg between the atmosphere and the soil (Yang et al. 2018) .

The studies of the Hg cycle in forest ecosystems show that gaseous elemental Hg(0) is the main source taken up by plants (Bishop et al. 2020; Zhou et al. 2021). Differences are observed between ecological functional traits and species among other factors. Hg in tree rings is also analyzed usually to determine if the Hg concentration are influenced more

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by the soil Hg concentration or by the atmospheric Hg deposition. This atmospheric deposition that has declined in recent decades can be shown in the declining concentration from the older to newer tree rings suggesting a more important uptake through the foliage than the roots. Hg dendrochemistry can help investigate Hg past impacts on terrestrial ecosystems and also help predict future changes in the cycle of Hg in forests (Yanai et al., 2020). The recent studies on Hg uptake by vegetation have highlighted the importance of the role of different parameters as vapor pressure deficit, soil water content, climatic conditions, date of sampling, leaf mass area, tree functional groups, stomatal conductance affecting potentially the root uptake of Hg dissolved in soil water and the absorption rate via stomata and eventually the Hg leaf content (Blackwell & Driscoll, 2015; Rea et al., 2002; Wohlgemuth et al., 2021; Yang et al., 2018b).

Analysis of long term atmospheric Hg(0) and CO_2 concentrations are very informative to understand the role of the vegetation in the global Hg cycle (Jiskra et al. 2018). Studies conducted in temperate Northern Hemisphere (44°N) show strong atmospheric Hg(0) seasonal variation, with high values in winter and low values in summer (Jiskra et al. 2018). These authors show also a significant positive correlation between the monthly Hg(0) and CO_2 concentrations. They highlighted a one-month offset in Hg(0) summer time minima happening in September in comparison to the CO_2 minima value occurring in August, this trend is not observed in winter time. The uptake of Hg(0) by the vegetation continues during CO_2 respiration periods during the fall and night when the ecosystem exchange of CO_2 turns from being a sink to becoming a source (Jiskra et al. 2018; Wofsy et al. 1993).

The total gaseous Hg (TGM) in the Mediterranean atmosphere is similar to Northern Europe (1.3 to 2.4 ng m^{-3}) (Kotnik et al. 2014). In the case of a semi-closed sea such as the Mediterranean basin with warm summers, high sea-water evaporation, solar radiations and Hg anthropogenic sources, the Mediterranean Sea is acting as a net source of Hg to the global atmosphere (Kotnik et al. 2014) making the Mediterranean an air-pollution area (Baayoun et al. 2019; Borjac et al. 2019).

The olive tree (*Olea europaea* L.) is one of the most distinctive Mediterranean agro-ecosystems tree species, and is adapted to drought (Sghaier et al. 2019). Considered among the oldest trees in the Mediterranean basin, these centennial olive trees are still growing in many countries along both the eastern and western shore, surviving to various stresses and witnessing the historical, cultural and ecological importance of this tree (Terral et al. 2004). On the other hand, the olive tree remains a key component of agriculture today and in the future. Therefore, it is important to understand the on-site behavior of the olive tree to Hg pollution in its natural Mediterranean growing environment. The Hg source in foliage varies with respect to the amount of contamination. In polluted sites the soil is the main source of Hg while away from those sites the atmosphere is the most important source (Naharro et al. 2018).

Lebanon, a small country at the Eastern Mediterranean, is facing important anthropogenic pressure within a changing environment (Gérard and Nehmé 2020). The air quality in Lebanon all over the country is noted to be moderately unsafe with an annual mean concentration of 31 $\mu g/m^3$ of PM 2.5 which is above the maximum recommended value (10 $\mu g/m^3$) (Lebanon: Air Pollution IAMAT 2020). Adding to that, soil samples collected from different areas in southern Lebanon showed values of Hg concentration ranging between 160-6480 ng/g (Borjac et al. 2020). The main contributors of the air pollution include cement industries, mineral and chemical factories, vehicles emissions, food

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processing and oil refining. Ancient olive groves are found across different agroclimatic areas at different altitudinal belts, still producing agricultural outcomes and consumed, coping with these various pollution pressures.

In this study, remote sites were investigated for possible Hg contamination that is expected to be very low, since the atmospheric Hg is still deposited in remote areas (Grigal, 2003). This is due to the surrounding polluted sites and the transfer of pollution through wind and the Mediterranean Sea to other areas. In addition to the lack and scarcity of studies on Hg pollution in the Eastern Mediterranean. The aim was to follow monthly concentrations variation of Hg during two consecutive years 2019-20 in the foliage and stems of olive trees, soils and litter of two ancient groves of Lebanon known to be over 1400 years old (Yazbeck et al., 2018). Specifically, two main objectives are taken into account: 1) How does Hg content vary seasonally in uncontaminated and remote sites? 2) Is Hg-uptake from the soil to the foliage low in uncontaminated sites?

2. Materials and methods

Two monumental olive groves were chosen in the context of their historical and agricultural importance, since these two sites are considered to contain olive trees of an age older than 1400 years old still providing till this date an agricultural outcome that is consumed.

2.1. Geographic setting and environmental context

2.1.1. Bchaaleh site (BC) - North Lebanon

This grove is situated in Batroun district (Latitude 34°12'06'' N, Longitude 35º49'23'' E, Altitude 1300 m.a.s.l.) (Figure 1a). Olive trees are growing rainfed in a sandy loam texture soil of grain size analysis of sand, silt and clay percentages are 52.8 %, 38.7 % and 10.7 % respectively. Soil pH is 7.07 ± 0.26 with organic matter and calcium carbonate contents are 1.7 % and 38.3 % respectively (Yazbeck et al. 2018). In this study, soil profiles of carbon and nitrogen contents were analyzed. Organic carbon contents decreased with soil depth from about 4 % at 0-1 cm (Soil surface) to 2.7 % at 30-60 cm. The total nitrogen is about 0.3 % at 1cm depth and 0.2 % at 30-60 cm depth. The olive trees are located on two terraces. The first terrace is at 1.5 meter above the road level while the second is at the road level. They are maintained by the municipality since the last four decades as an endowment property. Precipitation average ranges between 229 and 392 mm/year in winter and between zero and less than 2 mm/year in summer, while average temperature is between 4 and 8 °C in winter and between 20 and 23 °C in summer (data extracted from LARI climatic data) (Figure 1b, Table S1).

The village is at about 36 km from Chekka town located at a lower altitude (0-200 m.a.s.l.) nearby the sea (Figure 1a), and which is classified as a source of air pollution (EJOLT 2019). Chekka hosts an important national cement factory responsible of carbon dioxide, sulfur dioxide, nitrous oxides, monoxide and particulate material emissions causing respiratory and health issues (Kobrossi et al. 2002) and water pollution (Nassif et al. 2016). At 28 km from BC, the small commercial port of Selaata (0-37 m.a.s.l.) that emits many pollutants (ie. Phosphogypssum, heavy metals, radionuclides) expanded via water and air pathways (Petrlik et al. 2013; Yammine et al. 2010). As dissolved gaseous

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Hg from natural and human activities is saturated in the upper Eastern Mediterranean Sea, Gårdfeldt et al., (2003) have evidenced that Mediterranean Sea is a source of airborne elemental Hg.

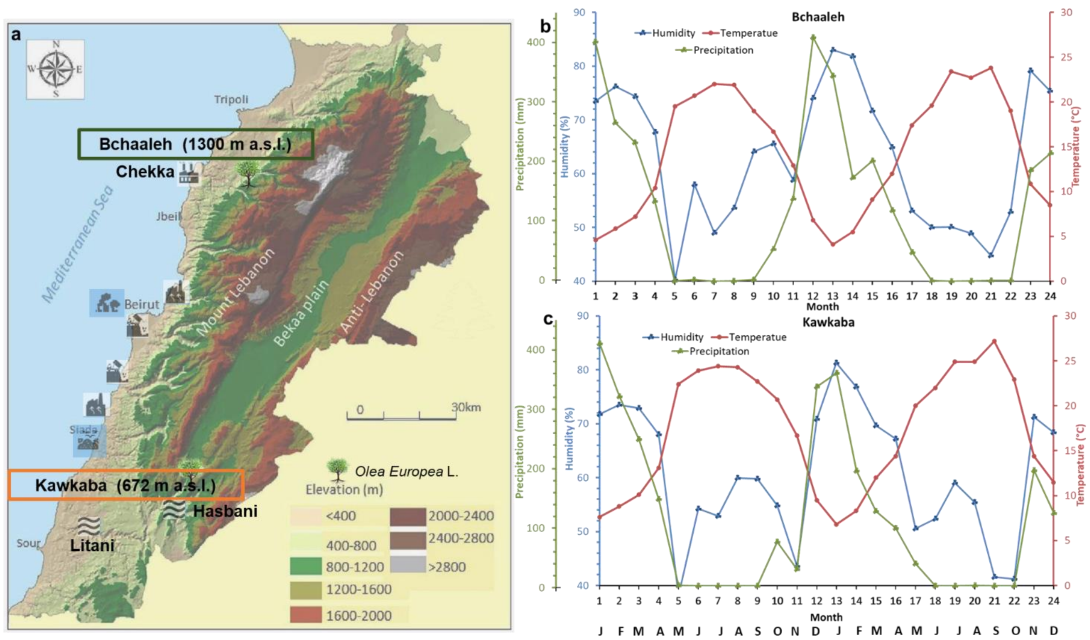

Figure 1. (a) Site locations of the two selected focus areas (modified after Shared Water Resources of Lebanon, Nova Science Publishers 2017). **(b,c)** Climatic data of Precipitation, Temperature and Humidity collected from the meteostations installed by LARI between 2009 and 2018 (Colors should be used for this figure in print)

2.1.2. Kawkaba site (KW) - South Lebanon

The second grove is located in the village of Kawkaba (KW), South Lebanon (Latitude 33°23'856'' N, Longitude 35°38'588'' E, Altitude 672 m.a.s.l. (Figure 1a). KW soil is characterized as clay loam soil of pH 7.5 ±0.5. Soil organic material and calcium carbonate average are 1.7 % and 59.0 % respectively (Al-Zubaidi et al. 2011) and grain size analysis of sand, silt and clay percentages are 6 %, 28 % and 66 % respectively. The analysis of organic carbon and nitrogen at the 0-1 cm and at 1-30 cm decrease from about 9.0 % to 2.2 % and from 0.9 % to 0.3 % for the carbon and nitrogen respectively. Average precipitation ranges between 215 and 374 mm/year in winter and drop to almost zero mm in summer, while average temperature is between 7 and 11 °C in winter and between 21 and 27 °C in summer (data extracted from LARI climatic data) (Figure 1c, Table S1).

The village has to its east the Hasbani river, originated from the north-western slopes of Mount Hermon in Hasbaya (36 km away from KW and located at 750 m.a.s.l.) (Badr et al. 2014; Jurdi et al. 2002). On the other hand, the Litani River (170 km long and located at 800 to 1000 m.a.s.l) (Figure 1a) rising in the south of the Bekaa valley is about 29 km away from KW (Abou Habib et al. 2015, Khatib et al. 2018). These two rivers are polluted and for these reasons

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they are not used for irrigating crops in KW and surrounding areas while the olive trees are growing rainfed as per indicated by the municipality of KW.

Climatic data in both BC and KW were collected from meteorological station and manual rain gauge installed in the villages by LARI (Lebanese Agricultural Research Institute) (Figure 1b,c). CO_2 data used in this study are from NOAA Global Monitoring Laboratory (https://gml.noaa.gov/webdata/ccgg/trends/co2/co2_trend_gl.txt).

2.2. Field sampling

For the Hg concentration analysis, four olive trees of 8 to 15 m foot circumference (and the average height of the trees 4-6m) were sampled in each of the two groves, within BC two trees taken in an upper terrace (BCO1-Bchaaleh-Tree 1, BCO4-Bchaaleh-Tree 4) and two other trees in a 1.5 m lower terrace (BCO9-Bchaaleh-Tree 9, BCO12-Bchaaleh-Tree 12) (Figure S1). For Hg analysis, foliage, stems and fruits were collected from the trees, while litter and soil were sampled directly under the trees, starting from February 2019 and ending by September 2020. For each olive tree, both sun exposed and shaded foliage (olive tree bears foliage from three different years) and stems (terminal portions of 20 cm) with no evidence of pathogens were randomly taken and merged from the upper, middle, and lower canopy position of the olive trees on a monthly basis using a manual pruner. Litter and soil surface were separately collected on the whole top surface area of the olive groves and stored in different paper bags once every four months. In parallel, soil sampling was performed using a bucket auger to a maximum depth of 60cm. These soil samples were collected from around the whole olive grove in order to be a good representative of the whole site. In both sites, the soil showed uniform color and texture. Soil cores were fractioned in soil surface (0-1 cm), 1 to 30 cm depth and from 30 to 60 cm depth in order to study the effect and accumulation of Hg concentration on the different depth layers. To avoid contamination, gloves were worn while collecting samples, and the equipment was rinsed with methanol between every sample. A set of 453 samples were collected and stored in paper bags until further preparation for the Hg analysis.

2.3. Sample preparation for Hg analysis

Collected foliage and stems were rinsed with distilled water and then dried for 48 hours in an oven at a temperature of 60°C at maximum. The dried foliage, stems, litter and olive fruits samples were grinded using an electrical stainless grinding machine with no heating system for 5-10 minutes, while soil samples were prepared with a manual natural agate grinder. All samples were later sieved using an inox stainless-steel 125-micron sieve mesh to collect homogeneous powders for analysis. A total of 150 mg for foliage and soil (50 mg/analysis), and 300 mg of litter and stems (100 mg/analysis) were considered in triplicates for analysis of Hg concentrations.

2.4. Analytical method

For the Hg elemental analysis, a total of 453 powder samples from foliage, stem, grain, litter and soil were analyzed using an advanced Hg analyzer AMA 254 (Altec) as described elsewhere (Barre et al. 2018; Duval et al. 2020). A

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known amount of sample (50-100 mg) is weight in a nickel boat, using a 10^{-6} g precision balance. The sample aliquot is first dried at 120°C for 60s and subsequently pyrolyzed at 750°C for 150s, under oxygen flow. The resulting gaseous Hg produced during the sample decomposition is amalgamated on a gold trap and then released to an Atomic Absorption spectrometer after a thermal desorption step at 950°C. The AMA 254 instrument was calibrated through several external matrix-matched calibration procedures using the following certified reference materials: IAEA-456 sediment (77 ± 5 ng Hg/g), NIST-1575A pine needles (39,9 ± 0,7 ng Hg/g) and IAEA336 (200 ± 40 ng Hg/g). The QA/QC evaluation of the analytical procedure was completed with a continuous monitoring of the blank's values (Nickel boat Hg background noise), every 15 analyzed samples. The precision of the measurements was assessed through replicated analyses (n=2) of 13 % of the total amount of samples (n=453). Average relative standard deviations of 5 % and 2.5 % are thus associated to the reported Hg concentrations for the 2019 and 2020 samples batches, respectively. The detection limit of the analytical method has been assessed to 0,7 ng Hg/g, for analytical sessions.
Susbamples of soil were used for carbon and nitrogen elemental contents (%) analysis. A 2 mg (acid washed soil and bulk soil) of powders were weighed into tin capsules and measured by dry combustion using a Pyrocube Elemental Analyser (EA, Elementar GmbH).

2.5. Statistical analysis

R 4.1.0 program was used for the statistical analysis, the data in not normally distributed. For the effect of tissue type on Hg concentration, Wilcoxon test was used with the tissue type (foliage and stems) as the main effect. Pearson correlation analysis was used to examine the inter-individual correlation of Hg concentration between the trees. Correlation between Hg concentration of soil surface, litter and foliage was studied using a correlation test. For the seasonal effect (Winter: Mid December till Mid-March, Spring: Mid-March till Mid-June, Summer: Mid-June till Mid-September, Autumn: Mid-September till Mid-December) on Hg concentration, Wilcoxon test was used considering the unequal data available for the different seasons. Finally, the effect of climatic factors (Temperature, precipitation, pCO_2) on Hg accumulation was examined using a Wilcoxon test.

3. Results

3.1. Hg concentrations in plant tissues, litter and soil at BC and KW groves

Hg concentrations varied generally according to both tree tissues and groves agroclimatic conditions (Table 1, Figure 1). Hg values in the foliage varied significantly between the two groves (p-value=$1.581*10^{-6}$), where the highest concentration was recorded in BC (48.1 ± 10.6 ng/g) *vs.* (35 ± 12.4 ng/g) in KW. Soil surface also recorded a difference in Hg concentration between BC and KW, with 61.9 ± 20.0 ng/g in BC and 128.5 ± 9.4 ng/g in KW. Soil 0-30 cm samples taken from BC and KW groves, values ranged between 31.8 ± 4.7 ng/g and 70.2 ± 23.4 ng/g respectively. In soil 30-60 cm Hg concentrations recorded 19.5 ± 6.73 ng/g at BC. No significant differences were recorded for litter and stems Hg concentrations (p-value= 0.0915 and p-value=0.2215 respectively) between the groves, with litter values of 62.9 ± 17.8 at BC and 75.7 ± 20.3 ng/g at KW *vs.* stem values of 7.9 ± 2.8 ng/g at BC and 9.0 ± 4.7 ng/g at KW.

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Positive correlations were observed between soil and litter in BC (r=0.60) and KW (r=0.95) though statistically insignificant (p-value= 0.40 and 0.13 respectively).

In descending order of Hg concentrations and considering the different sites, plant tissue, soil and litter samples, the Hg concentrations could be ranked in BC, soil surface > litter> foliage > soil 0-30 cm > soil 30-60 cm > stems; and in KW, soil surface > litter > soil 0-30 > foliage > soil 30-60 > stems (Table 1).

Table 1. Overall mean values of Hg concentration (ng/g) of the different studied material in both BC and KW olive groves

	Bchaaleh (BC)			**Kawkaba (KW)**		
	Average (ng/g)	**SD**	**N**	**Average (ng/g)**	**SD**	**N**
Foliage	**48.1**	**10.6**	**66**	**35.0**	**12.4**	**67**
Stems	**7.9**	**2.8**	**66**	**9.0**	**4.7**	**67**
Litter	**62.9**	**17.8**	**7**	**75.7**	**20.3**	**6**
Soil Surface	**61.9**	**20.0**	**8**	**128. 5**	**9.4**	**6**
Soil 0-30cm	**31.8**	**4.7**	**6**	**70.2**	**23.4**	**5**
Soil 30-60cm	**19.5**	**6.7**	**5**	**28.0**		**1**
Fruit	**7.0**	**3.5**	**3**	**11.0**		**1**

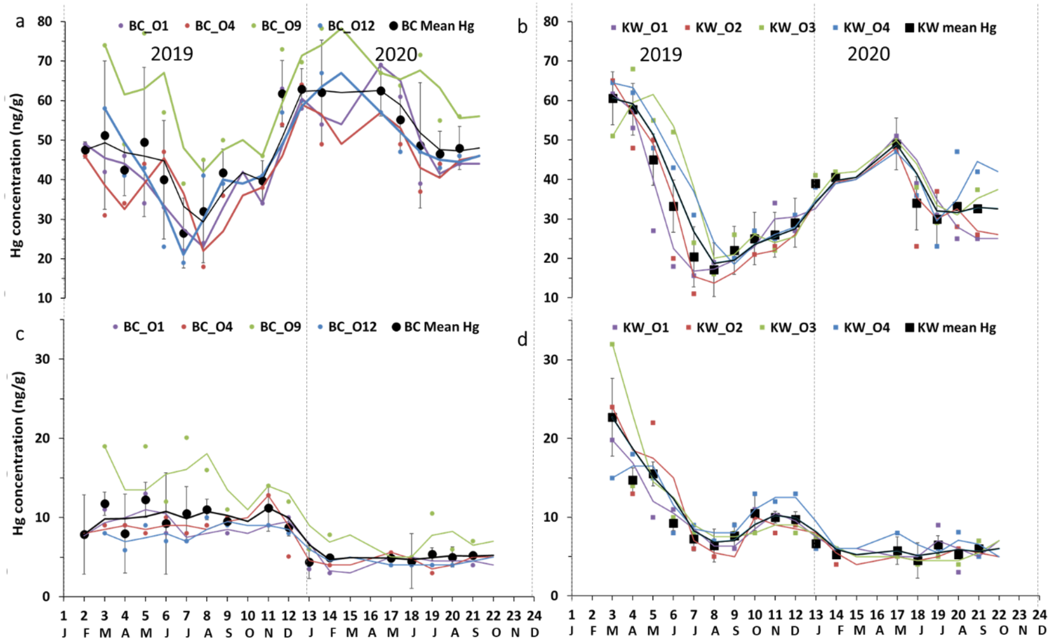

Figure 2. Seasonal variations of foliage Hg concentration in**(a)** BC and **(b)** KW olive groves and stems Hg concentration in **(c)** BC and **(d)** KW olive groves.

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3.2. Seasonal variation of Hg concentration

Hg concentrations recorded between February 2019 and September 2020 reflected a significant seasonal variation in both sites (p-value<$2.2*10^{-16}$). In BC grove, foliage registered its highest Hg concentration during winter and spring with 61.8 ± 7.6 ng/g and 55.1 ± 12.5 ng/g respectively, and its lowest Hg amount during summer and autumn with 41.5±12.7 ng/g and 44.4 ± 6.2 ng/g, respectively. Seasonal effect on foliage and stems (p-value<$2.2*10^{-16}$), the lowest Hg concentration values for all the mentioned samples were registered during winter. The stems and soil 0-30cm highest values was registered in autumn (Table 2). Significant differences were found in foliage Hg values between summer and winter (p-value=0.00020), and autumn and winter (p-value= 0.00014). Similarly, stems Hg values varied significantly between spring and winter (p-value=0.030), autumn and winter (p-value=0.047). In KW, the highest Hg concentrations for foliage and stems were registered in spring with 51.8 ± 4.5 ng/g, 11.7 ± 6.7 ng/g respectively. Significant differences were found in foliage Hg values between summer and winter (p-value=0.013), autumn and winter (p-value= 0.00067), autumn and spring (p-value=$1.589*10^{-05}$), spring and winter (p-value= $9.383*10^{-05}$) and spring and summer (p-value=$2.327*10^{-06}$). Similarly, stems Hg values varied significantly between spring and winter (p-value=0.006), spring and summer (p-value=0.0036) and autumn and spring (p-value=0.011). A seasonal variation is observed in both olive groves especially in the foliage and also between the two groves location.

Table 2. Seasonal mean Hg concentration (ng/g) and standard deviations seasonal variation of the different studied material in both BC and KW olive groves. Grey color indicated the highest Hg concentration values among the different elements during the different seasons

Hg (ng/g)												
Bchaaleh	**Spring**	**SD**	**N**	**Summer**	**SD**	**N**	**Autumn**	**SD**	**N**	**Winter**	**SD**	**N**
Foliage	55.1	12.5	16	41.5	12.7	24	44.4	6.2	12	61.8	7.6	18
Stems	7.8	3.8	16	7.61	3.9	24	8.3	2.7	12	6.4	2.9	18
Litter	79.3	26.5	3	64.7	4	4	55.5		2	48.6	13.3	3
Soil Surface	58.3	13	3	84.5	21.2		50		3	50.6	23.5	3
0-30cm	33.6	6.2	2	32.2	4.3		34.5		2	27	0.7	3
30-60cm	23.1	9.1	2	20.7			19.6		2	11		1
Kawkaba	**Spring**	**SD**	**N**	**Summer**	**SD**	**N**	**Autumn**	**SD**	**N**	**Winter**	**SD**	**N**
Foliage	51.8	4.5	16	28	7.2	24	28.5	7.2	16	33.9	5.6	18
Stems	11.7	6.7	16	6.5	1.4	24	7.7	2.1	16	6.9	1.6	18
Litter	90.1	29.3	2	67	24	2	70	1.4	2			
Soil Surface	132	8.5	2	118	4.2	2	135.6	2.2	2			
0-30cm	57.9	11.2	2	65.8		1	84.8	36.4	2			
30-60cm	28		1									

3.3. Tree comparison

In the upper terrace of BC grove, the foliage and stems average Hg concentration per tree ranked respectively between 44.6 ± 13.3 ng/g and 7.1 ± 2.9 ng/g for BCO1 and between 42.4 ± 11.5 ng/g and 7.0 ± 2.8 ng/g for BCO4. In the lower terrace of the same site, Hg concentrations were found respectively between 60.7 ± 12.7 ng/g and 11.2 ± 5.2 ng/g for BCO9 and between 45.6 ± 12.7 ng/g and 6.4 ± 2.2 ng/g for BCO12 (Figure 2a,c).

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In KW grove, trees show no significant difference for both foliage and stems. The average concentration per tree in foliage and stems were 32.4 ± 12.2 ng/g and 8.5 ± 4.0 ng/g respectively for KWO1, 32. 8 ± 14.7 ng/g and 8.9 ± 6.0 ng/g for KWO2, 37.6 ± 14.0 ng/g and 9.3 ± 6.7 ng/g in KWO3 and 37.7 ± 13.6 ng/g and 9.6 ± 4.0 ng/g in KWO4 (Figure 2b,d).

In BC grove, the trees located on the lower terrace recorded higher Hg concentration values than those of the upper terrace especially tree 9. While KW grove had similar Hg concentration among all four trees.

3.4. Hg concentration and agro-climatic effect

The Wilcoxon test for a non-normal distribution between Hg concentration of foliage and stems and temperature shows no significant effect with a 0.013 p-value. While a significant effect was found between Hg concentration of foliage and stems, precipitation, relative humidity and atmospheric CO_2 (pCO_2) (p-value $2.2*10^{-16}$, $5.60*10^{-10}$ and 2 $.2*10^{16}$ respectively). A positive correlation was found between the precipitation and Hg concentrations of foliage and stems, where values increased with higher precipitation and lower temperature records. In dryer seasons, starting from May to mid-October, the Hg concentration of foliage decreased with the increase of temperature and the decrease of the precipitation for all studied elements.

4. Discussion

4.1. Hg concentration in plant tissues, soil and litter in the studied groves

In both groves our values showed a larger Hg concentration in the olive foliage (BC average of 48.1 ± 10.6 ng/g; KW average of 35.0 ± 12.4 ng/g), than that of stems (BC average of 7.9 ± 2.8 ng/g; KW average of 9.0 ± 4.7 ng/g) and that of olive fruits (7 ± 3.5 ng/g at BC, n=3 and 11 ng/g in KW, n=1). Our data corroborates previous studies (Bargagli 1995; Higueras et al. 2016; Naharro et al. 2018) showing that olive foliage has the main Hg content among analyzed plant tissue. Moreover, we demonstrate that our values are lower than 200 ng/g considered as Hg pollution threshold (Kabata-Pendias and Pendias 2000) and confirm no pollution effect for both BC and KW groves (Table S2; Figure S2a,b). This been said, our sites are good remote bioindicators of the uptake of Hg through the plant although more prolonged time range is needed. Hg in foliage originates predominantly from the air, and it can be delivered to the soil through the litter (Tomiyaso et al. 2003). Adding to that, the atmospheric Hg uptake in foliage dominates over Hg release (Pleijel et al., 2021). Since normally the main source of Hg into the foliage is atmospheric and minimally through the soil, this explains the higher Hg concentration in the foliage, while in the stems the main uptake is from the soil and it seems to be minimal (Tomiyasu et al., 2005) explaining the much lower level of Hg concentration in the stems than that in the foliage.

The soil surface and litter registered the highest Hg concentration (62 to 129 ng/g) among all samples (foliage, stems, fruit) in both groves (Table 1) suggesting that the soil is the main Hg reservoir through the Hg throughfall and litter inputs. Our findings are in agreement with studies on evergreen forest ecosystems reporting that soil can hold more than 60 % of Hg input to the forest floor (Wang et al. 2016). Our soil surface sites values (61.9 ± 20.0 ng/g in BC and

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128.5 ± 9.4 ng/g in KW) show higher Hg concentration in KW compared to the general background level of Hg as defined by uncontaminated soil world reference mean Hg contents (20 to 100 ng/g; Kabata-Pendias and Pendias 2000; Senesil et al. 1999; Gworek et al. 2020). However, both sites have significantly lower values compared to known industrial and mining contaminated sites (> 1000 ng/g ; Kabata-Pendias and Pendias 2000; Higueras et al. 2016). Nevertheless, studies conducted in different sites show a wide range of natural background Hg levels (ie. topsoils in Europe, India, Brazil, Norwegian Arctic, New Zealand have values of 40, 50, 80, 110, 230 ng/g respectively (Gworek et al. 2020) making it difficult to set a specific Hg threshold value for uncontaminated soil (Table S2; Figure S2c). Due to the differences registered in different countries and sites of sampled soil, this indicates a link with chemical and mineralogical soil properties (ie. pH, humic acid, soil grain size distribution, organic matter type and clay percentage) affecting Hg in soil and its transport (O'Connor et al. 2019). Hence, we suggest that lower values in BC soils are likely explained by the low clay, organic carbon and nitrogen contents (10.7 %, 4 % and 0.3 % in soil surface respectively). While KW higher Hg soil contents can be explained by the higher clay proportion (66 %) and organic carbon and nitrogen contents (9 % and 0.92 %). On such clay loam soils and rich organic matter, Hg binding is facilitated explaining higher content (O'Connor et al. 2019). Moreover, the higher temperatures and lower precipitation and altitude in KW can be responsible for a higher dry deposition from gaseous elementary Hg in the forest floor (Teixeira et al. 2017).

In parallel, the litter showed higher Hg concentration than that in foliage in both BC (62.9 ± 17.8 ng/g) and KW (75.7 ± 20.3 ng/g) (Table 1) likely explained by the process of the Hg input into the litter through foliage shedding and throughfall to the forest floor (Rea et al. 1996; Pleijel et al. 2021).

4.2. Seasonal foliage Hg content versus seasonal atmospheric Hg and CO_2

The late winter-early spring registered the highest Hg concentration for foliage in both groves, while summer and early fall to a less extent recorded the lowest concentrations (Figure 1a,b). Unexpectedly, we found a good positive covariation between Hg contents of our olive tree foliage and that of the atmosphere in the Northern Hemisphere (Jiskra et al. 2018). According to (Obrist, 2007), the observed maximum Hg_{atm} (from October to March) can be explained by the anthropogenic and natural Hg emissions (ie. soil carbon mineralization, plant dormancy, net respiration of the global forest ecosystems) exceeding Hg burial processes (ie. Photosynthetic activity and stomatal conductance, forest floor Hg uptakes).

This author discussed the significant role of the northern hemisphere vegetation and forest floor respiration and suggested that the decreasing atmospheric Hg content starting in March and reaching the minimal values in July-August is likely attributable to the plant uptakes (Obrist 2007; Jiskra et al. 2018). However at our latitudes, BC and KW foliage evergreen olive trees show also a decrease in Hg contents from end of March to late September, with minimum values centered in August suggesting a decline of the plant Hg uptake likely explained by the reduction of the stomatal conductance (Lindberg et al. 2007; Pleijel et al. 2021). This minimal photosynthetic activity occurs during the driest (0 mm precipitation) and hottest temperatures (above 25°C) in our sites. While the increase of the Hg intake starting in September and reaching its maximum in March occurs during the increasing Northern Hemisphere atmospheric Hg (Figure 3a,b). In different Olive cultivars from Italy, Proietti and Famiani (2002) show that lowest photosynthetic activity occur during summer (August) but also in winter (December) which is not supported by our data. The positive covariation between our olive foliage and atmospheric Hg contents do not support the previous cited studies showing a strong depletion of atmospheric Hg concomitant to vegetation uptake in summer period uptakes (Obrist 2007; Jiskra et al. 2018). Two hypotheses can explain such positive correlations between our plant foliage and air Hg concentrations. Taking into account Hg results obtained during several seasonal campaigns in the surface waters of the Mediterranean Sea and the above atmospheric layer, it has been evidenced that the highest Hg fluxes to the atmosphere occur mainly in summer (Kotnik et al. 2014; Wangberg et al. 2001) and a higher TGM is registered in the eastern Mediterranean compared with the western Mediterranean and Northern Europe (Wängberg et al., 2001). Hence, we can hypothesis that a reverse Hg seasonal cycle compared to the Northern Hemisphere values would stand on a different seasonal atmospheric Hg variation than in the Northern Hemisphere.

On the other hand, other studies reported the same positive correlation between atmospheric Hg and crops (Niu et al. 2011). This suggest a second hypothesis where our groves seasonally exposed to high atmospheric Hg, accumulate Hg in their foliage (Lindberg et al. 2007; Pleijel et al. 2021). According to Hanson et al. (1995), a compensation point for Hg uptake by plant foliage can be considered but no information to our knowledge is available for the specific case of the olive trees. The tight link between foliage Hg uptake and stomatal conductance seasonal variations can be also deduced from the analysis of the partial pressure of the pCO_{2atm} seasonal variation (Obrist 2007; Sprovieri et al. 2016; Jiskra et al. 2018; Obrist et al. 2018; Pleijel et al. 2021) (Figure 3b,e,f). Very good covariation between olive foliage Hg and pCO_{2atm} are shown for BC and KW despite a notable offset of one month at KW to two months at BC can be deduced (Figure S3). Taking into account our calculated time lags, we obtained significant correlations between our foliage Hg content and pCO_{2atm} of 0.718 and 0.704 in BC and KW respectively. Interestingly a one month time-lag between atmospheric Hg and pCO_{2atm} is also reported by Jiskra et al. (2018) for most northern hemisphere sites. The offset of one to two months between maxima of BC and KW foliage Hg (March/April) and pCO_{2atm} (May) suggests that the minimum of Hg in the foliage occur during the decreasing phase of the pCO_{2atm} when the global northern hemisphere tend to become a net sink of CO_2. When minimum values of pCO_{2atm} are reached at the end of the dry summer (Figure 3b), concomitant to minimum atmospheric Hg (Figure 3a), end of the drought and increase of precipitation (Figure 3c, d), BC and KW olive trees show a rerise of the Hg uptake. The photosynthetic activity and the stomatal conductance related to the climatic parameters (temperature, precipitation, humidity, pCO_2) as shown by Ozturk et al. (2021) and the atmospheric Hg explain our foliage Hg seasonal cycle. At a regional scale, our sites show different time lags between BC and KW that we cannot explain fully except their altitudinal differences which can suggest that BC grove benefits of less drought in summer. This can also be explained by the physiology of olive trees that is characterised by small foliage surface area as a response to the high temperature and less humidity in summer and the stomatal conductance in the foliage that increase in winter and decrease in summer. As per (Connor & Fereres, 2010), olive is a day neutral plant were its reproductivity is affected by temperature and sunlight. Its vegetative growth is limited by the low temperature in the winter season and water supply in the summer season. The water loss from the foliage due to transpiration and temperature, creates a replacement for water source to be the soil through the roots. The xylem being the main transporter between the roots and the canopy. During the day the evaporation increase and the plant water content decreases and become minimal during midday, noting that the soil water content is high.

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During the evening the olive tree seeks soil water as an equilibrium. If soil water is not available enough to cover the olive need, the evergreen olive is able to adapt through conservation of internal water especially during severe summer drought, due to its ability to restrict water loss to the atmosphere and maximize extraction of soil water. Further analysis and data are necessary to confirm the above assumption.

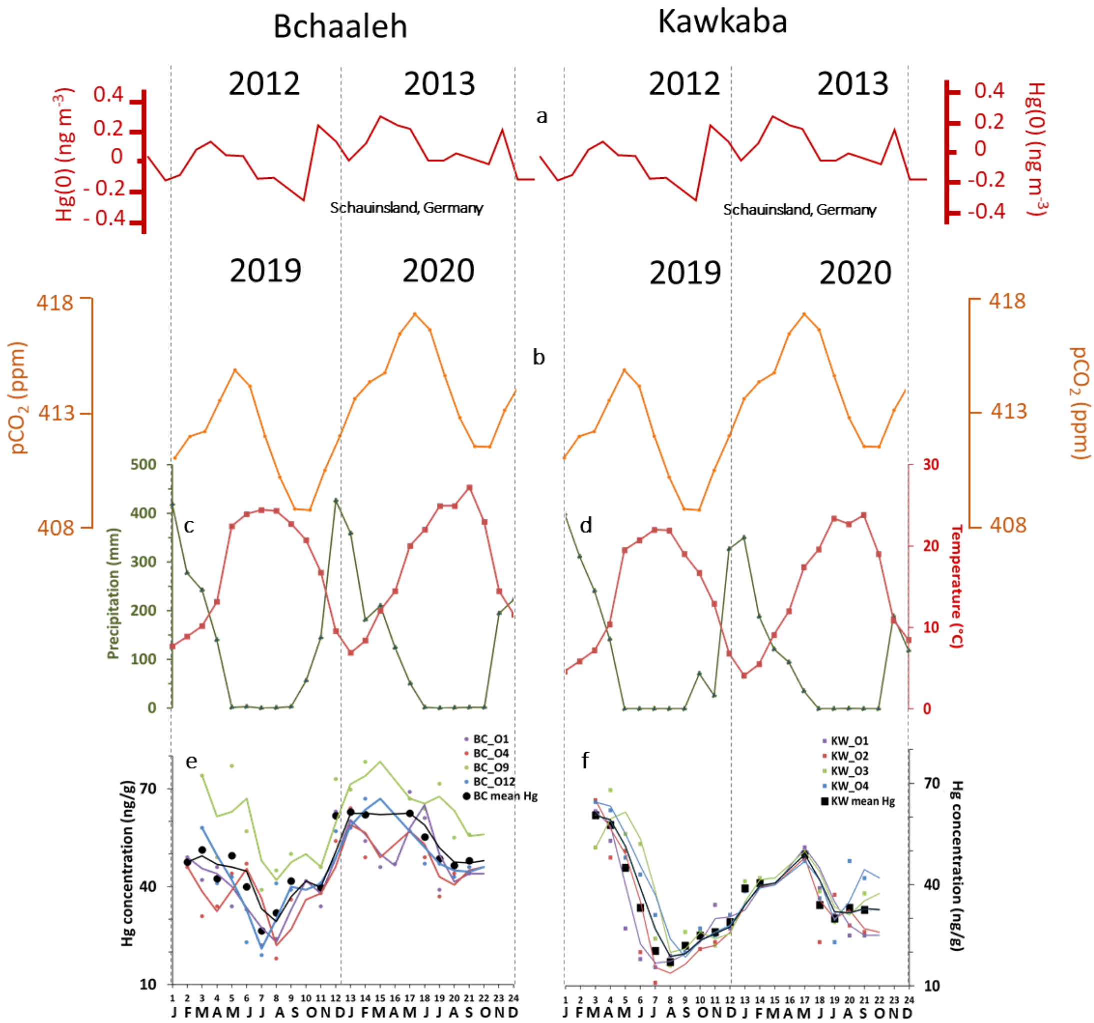

Figure 3. Seasonal variations of **(a)** atmospheric Hg(0) (Jiskra et al., 2018), **(b)** pCO_2 (NOAA Global Monitoring Laboratory), **(c)** and **(d)** precipitations and temperature of BC and KW respectively and **(e)** and **(f)** foliage Hg concentration in BC and KW olive groves respectively

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4.3. Hg uptake in BC and KW grove systems

4.3.1. Hg cycling in the stems, litter and soil system

For each site, Hg contents in stems exhibit narrow range between the different trees except BCO9 tree showing the highest values as observed for the foliage. We speculate that this higher Hg content is the adjunction of chemical products as fertilizer on the plot 549 likely between fall and winter (Figure S1) belonging to different owner.
At a seasonal scale, the averaged Hg values of stems, litter and soil system show no statistical significance differences between the four seasons for the litter and soil while stems show a significant difference between winter (lowest values) and spring (p-value= 0.030) and winter-autumn (p-value= 0.047) in BC grove mostly similar to foliage changes. The same behavior was registered in KW in litter and soils, while stems showing statistical significance differences between autumn and spring (p-value= 0.011), spring-winter (p-value= 0.006) and spring-summer (p-value= 0.004) (Table 2). Despite the small amount of Hg content in the stems, the statistically significant seasonal changes may suggest that small amount of Hg move from the foliage to the lignified tissues as stems. However, we cannot neglect the Hg transport in xylem sap from the roots to the aboveground plant tissues even if minimal (Yang et al. 2018).
We can suggest the following Hg cycling in the system of the Olive grove/soil. In winter-early spring the highest concentrations in foliage feed continuously the litter and can explain the following maximal spring Hg content in the litter. The decomposition of the litter organic matter during the wettest conditions likely liberate Hg in the Hg(0) or Hg(II) forms or MeHg either towards the atmosphere or the surface soil respectively. A fraction of the degraded organic matter is transferred through gaseous evaporative processes towards the atmosphere while another fraction of the Hg is lixiviated towards the deeper soil in addition to dry Hg deposition during dry season (Teixeira et al. 2017). We can also speculate that the small Hg decrease observed during the winter season in BC can be due to the transport of total Hg and MeHg through the roots and xylem sap to the above ground tissues.

4.3.2. Hg uptake in foliage: atmospheric or soil sources?

In uncontaminated sites as those presented in this study, no significant correlation between the foliage and soil Hg content is observed in agreement with Higueras et al. (2012). In contrary, contaminated sites, exhibit a good correlation (Higueras et al. 2016). Interestingly, in the Mediterranean region, it is reported that intensity of mycorrhizal colonization of olive tree roots increase with the increase of seasonal precipitation and decreased with the increase of air temperature (Meddad-Hamza et al. 2017). This is an independent argument to support the atmosphere being the main Hg source to the foliage during the wet seasons, but also confirming in absence of mycorrhizal activity a possible minimal Hg uptake from the soil to the foliage and stems through water during the drought season (Rea et al. 2002; Meddad-Hamza et al. 2017). Eventually, we suggest that the main source of Hg for both olive groves is the atmospheric mercury.

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5. Conclusion

This is the first study conducted on monumental olive trees in a non-contaminated site of the MENA region and followed at a monthly basis over 18 months. Findings of our study in remote and uncontaminated sites indicate a higher uptake of Hg in the olive foliage compared to stems, fruits items and a remarkable Hg_F seasonal variation in both studied groves. Winter and Spring were particularly suitable for Hg accumulation in foliage in both sites. The significant correlation between our foliage Hg contents and the atmospheric Hg content and pCO_2, despite the one to two months' time lag, suggests that 1) the main source of foliage Hg is the atmospheric Hg 2) the main factor explaining the seasonal Hg accumulation in foliage is due to the photosynthetic activity and stomatal conductance, but it can also be due to the physiology of olive trees. Thus, a more intensive study on the physiology of olive tree must be focused on, and more intensive studies on foliage, soil and litter is needed to be able to assess the source of Hg uptake in the olive trees. However further comparison and studies on the seasonal atmospheric Hg in the eastern Mediterranean basin are necessary to test our hypothesis of the reversed seasonality of Hg since contrary to the global Northern Hemisphere vegetation, our olive groves act as a sink of Hg and CO_2 when global Northern vegetation is emitting and vice-versa. This relationship Hg_F – Hg_{atm}-pCO_{2atm} should be further investigated along the season and locally to better understand the observed time lags. Soil surface registered the highest Hg concentration among all studied compartments due to well-known processes of litter and throughfall. Moreover, this study highlights significant differences between Hg soil groves due to differences in soil characteristics. In this study we worked on the present in order to have a better understanding of the Hg cycle in the olive tree. A second step would be to reconstruct the paleo-evolution through also studying tree rings Hg concentration were we can also identify a seasonal variation. Our main contribution in this study is to see how the present-day olive trees records some elements such as Hg to better understand the Hg in tree rings for the past.

Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Author Contributions

The corresponding author **Ilham Bentaleb** is responsible for ensuring that the descriptions are accurate and agreed upon by all authors. Conceptualization and methodology were done and developed by **Ilham Bentaleb** and **Lamis Chalak**. The material collection was performed by **Nagham Tabaja**, **Ilham Bentaleb**, **Lamis Chalak**, **Ihab Jomaa, and Milad Riachy**. Sample storage and preparation in Lebanon organized by **Nagham Tabaja**. Material preparation at ISEM by **Nagham Tabaja**, Data collection and analysis were performed by Nagham Tabaja, **Ilham Bentaleb**, **David Amouroux,** and **Emmanuel Tessier**. The setting of the meteorological stations by **Ihab Jomaa**. Subsamples of soil were analyzed for carbon and nitrogen elemental contents (%) by **François Fourel**. The first draft of the manuscript was written **by Nagham Tabaja**. **Ilham Bentaleb, Lamis Chalak**, **David Amouroux, Ihab Jomaa, and Milad Riachy** commented on previous versions of the manuscript. All authors read and approved the final manuscript. Supervision was done by Ilham Bentaleb and Lamis Chalak.

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors would like to acknowledge the National Council for Scientific Research of Lebanon (CNRS-L) and Montpellier University for granting a doctoral fellowship (CNRS-L/UM) to Nagham Tabaja. The authors would also like to thank the Franco-Lebanese Hubert Curien Partnership (PHC-CEDRE) project 44559PL for the funding provided. Institute of Evolutionary Science of Montpellier (ISEM) at Montpellier University and the Research Platform for Environment and Science- Doctoral School of Science and Technology (PRASE-EDST) at the Lebanese University are also acknowledged for their support of the laboratory work. Credits go to the Lebanese Agriculture Research Institute (LARI) for assuring automated weather stations and manual rain gauges per site. The authors are grateful to the Municipality of Bchaaleh (Mr. Rachid Geagea) and the Municipality of Kawkaba (Ms. Mira Khoury). Acknowledgments are extended to Ms. Amira Yousef at LARI for her kind support and to Mr. Akram Tabaja for helping during fieldwork.

Funding

This work was supported and funded by Lebanon and Montpellier University (CNRS-L/UM grant), and PHC-CEDRE-project 44559PL

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