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LEVEGŐSZENNYEZŐ KOMPONENSEK BIOAKKUMULÁCIÓJÁNAK ELEMZÉSE LABORATÓRIUMI KÖRÜLMÉNYEK KÖZÖTT

ANALYSIS OF BIOACCUMULATION OF AIR POLLUTANT COMPONENTS UNDER LABORATORY CONDITIONS

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ANALYSIS OF BIOACCUMULATION OF AIR POLLUTANT COMPONENTS UNDER LABORATORY CONDITIONS

Az értekezés doktori (PhD) fokozat elnyerése érdekében készült a Pannon Egyetem
i és Környezettudományi Doktori Iskolája keretében

Környezettudományok tudományágban

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Elfogadásra javaslom (igen / nem)

…….
(témavezető)

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Elfogadásra javaslom (igen / nem)

…….
(témavezető)

Az értekezést bírálóként elfogadásra javaslom:

Bíráló neve: …..... …..... igen /nem

…….
(bíráló)

Bíráló neve: …..... …..... igen /nem

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(bíráló)

A jelölt az értekezés nyilvános vitáján ….....%-ot ért el.

Veszprém, …….. …….
(a Bíráló Bizottság elnöke)

A doktori (PhD) oklevél minősítése….....
Veszprém , …….. …….
(az EDHT elnöke)

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Abbreviations

Ace – Acenaphthene

Acy - Acenaphthylene

Ant - Anthracene

B(a)a; - Benzo[a]anthracene

B(a)p – Benzo[a]pyrene

B(b)f – Benzo[b]fluoranthene

B(e)p – Benzo[e]pyrene

B(g,h,i,)p – Benzo[g,h,i]perylene

B(k)f – Benzo[k]fluoranthene

Cry – Chrysene

D(a,h)a – Dibenzo[a,h]anthracene

Flu – Fluorene

Fluoranthene- Flt;

GC-MS – gas chromatography mass spectrometry

HMW – high molecular weight

IP – Indeno[1,2,3-cd]pyrene

LMW - low molecular weight;

Me-Nap - 2-methyl-naphthalene

Methy-Nap - 1-methyl-naphthalene

Nap - Naphthalene

PAH - polycyclic aromatic hydrocarbon;

Phen – Phenanthrene

PM – particulate matter

PM0.1 - particulate matter 0.1 micrometers or less in diameter

PM10 - particulate matter 10 micrometers or less in diameter

PM2.5 - particulate matter 2.5 micrometers or less in diameter

Pyr– Pyrene

BFC – Bioconcentration factor

SIM - *Selective* ion monitoring

TIC - Total ion chromatogram

LOQ - Limit of Quantitation

LOD- Limit of detection

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Abstract

Due to the growing urbanization and the increase in the number of vehicles in Europe and worldwide, the quality of the air in big cities is increasingly being degraded by traffic-derived aerosol particles (particulate matter- PM). These airborne dust particles smaller than 10 micrometers and 2.5 micrometers (PM10 and PM2.5), as well as air pollutant components bound on their surface, pose a potential risk to plants. In addition to instrumental data collection the measurement of accumulated amount of potential harmful pollutants by plants could be used to classify the quality of the air.

The aim of my research was to investigate the bioaccumulation of air pollutant components under controlled (laboratory) conditions, using a standard protocol. My research work includes the examining the applicability of the 227.OECD standard for estimating bioaccumulation and determining the actual bioaccumulation capacity of selected plant material. Accordingly, I performed the following tests:

I. The accumulation of PAHs were investigated in lettuce (*Lactuca sativa* L.) under laboratory conditions. A total of 19 PAHs were found in the lettuce sample, of which 16 are priority PAHs classified by the US EPA. Among the EPA PAHs, the only exception was dibenzo(a,h)anthracene, which was not detected in our plant samples.

II. Bioaccumulation potential of 8 different vegetables were compared under controlled laboratory conditions. Striking differences were found in the accumulation potential of vegetables, which can only be partially explained by leaf morphological parameters. Probably, the different growth rates also affect the concentration of accumulated PAHs. The accumulation pattern of vegetables showed some similarities, with a predominance of lower molecular weight (LMW) PAH compounds for all plants.

III. Lettuce plants (*Lactuca sativa* L.) were placed in pots in small and medium-sized settlements in Veszprém County to test the level of air pollution with bioaccumulation tests. The accumulated PAH concentrations in lettuce plants ranged widely. Based on the results obtained the burning of biomass was the main source of PAHs, which included household heating and the burning of agricultural waste in the garden but transport-related emissions also contribute. The fact that both lettuce and soil samples showed the highest contamination in the

Hárskút Natura 2000 area draws our attention to the importance of analyzing individual pollution sources.

IV. The amount of accumulated PAHs in plantain (*Plantago lanceolate* L.) were determined in samples collected from different sampling sites and compared with sample treated under laboratory conditions. Possible sources were identified by using traditional source identification methods. The experiment proved that P. lanceolata is a reliable passive monitor for assessing the distribution pattern of PAH pollution. All samples were characterized by HMW PAH prevalence, in contrast to most reported studies. In addition, the experimental treatment under laboratory conditions served as a reference against which samples collected in the field could be compared.

V. Spider plant (*Chlorophtum comosum* L.) was used for indoor bioaccumulation tests. I found significant differences between the cooking practices and the accumulated quantities of PAHs. In households where the amount of used sunflower oil was higher, the relative proportion of HMW PAHs was significantly higher than in those where the use of lard and butter was dominant. The carcinogen B(a)p was measured in detectable amounts where baking was used as the most common cooking practice.

Based on the obtained results, it can be concluded that by using the protocol, important information can be collected about the accumulation pattern of the individual vegetables and clear dose-effect relationships can be drawn. However, field studies are needed to assess the actual risk of consuming contaminated vegetables.

Tartalmi összefoglaló

Az Európában és világszerte is tapasztalható növekvő mértékű városiasodás, a gépjárművek számának növekedése miatt a nagyvárosok levegőjének minőségét egyre növekvő mértékben rontják a közlekedési eredetű aeroszolrészecskék (szálló porok). Ezek a 10 mikrométernél illetve a 2,5 mikrométernél kisebb méretű szálló porszemcsék (PM10 illetve PM2.5), valamint felületükön megkötött levegőszennyező komponensek potenciális kockázatot jelentenek növények számára. A levegő minőségének vizsgálatára műszeres adatgyűjtés mellett alkalmazhatunk ún. passzív biomonitorokat, azaz a terhelés mértékét a növényekben akkumulált károsanyag tartalom alapján minősítjük.

Kutatásom célja volt a levegőszennyező komponensek bioakkumulációjának vizsgálata, ellenőrzött (laboratóriumi) körülmények között, szabványos protokoll alkalmazásával. A kutató munkám magában foglalja a 227. sz. OECD szabvány alkalmazhatóságának vizsgálatát bioakkumuláció becslésére, továbbá kiválasztott növényanyag tényleges bioakkumulációs kapacitásának meghatározását. Ennek megfelelően következők vizsgálatokat végeztem el:

I. laboratóriumi körülmények között vizsgáltam a PAH-ok aeroszolból történő felhalmozódását salátában (*Lactuca sativa* L.). Összesen 19 PAH-t találtunk a salátamintában, ebből 16 az US EPA által besorolt prioritást élvező PAH. Az EPA PAH-ok közül az egyetlen kivétel a dibenzo(a,h)antracén volt, amelyet növénymintáink nem mutattak ki.

II. 8 különböző zöldségféle bioakkumulációs potenciáljának hasonlítottam össze ellenőrzött, laboratóriumi körülmények között. A zöldségfélék akkumulációs potenciáljában szembetűnő különbségeket találtam, ami a levélmorfológiai paraméterekkel csak részben magyarázható. Valószínűleg az eltérő növekedési ütemek is befolyásolják a felhalmozódott PAH-k koncentrációját. A zöldségfélék akkumulációs mintázata némi hasonlóságot mutatott, minden növény esetében az alacsonyabb molekulatömegű (LMW) PAH-vegyületek túlsúlyával.

III. Veszprém megyei kis- és közepes településeken cserépben kihelyezett saláta tesztnövényt (*Lactuca sativa* L.) alkalmazva bioakkumulációs vizsgálatokkal felmértem a levegőszennyezettség mértékét. A salátanövényekben felhalmozódott PAH-koncentrációk széles tartományban mozogtak. A kapott eredmények alapján a biomassza égetése volt a PAH-ok legfőbb forrása, ami magába foglalta a háztartási fűtést és a mezőgazdasági hulladékok kerti égetését is. Emellett a közlekedéssel kapcsolatos kibocsátások is hozzájárulnak. Az a tény, hogy

mind a saláta, mind a talajminták a Hárskút Natura 2000 területen mutatták a legmagasabb szennyezettséget, felhívja a figyelmünket arra, hogy mennyire fontos az egyes szennyezőforrások elemzése.

IV. különböző karakterű mintavételi helyekről gyűjtött valamint laboratóriumi körülmények között kezelt útifű (*Plantago lanceolate* L.) mintákban határoztam meg az akkumulált PAH mennyiségét és elvégeztem a lehetséges szennyező források azonosítását hagyományos forráskijelölési módszerekkel. A kísérlet bizonyította, hogy a P. lanceolata megbízható passzív monitor a PAH-szennyezettség eloszlási mintázatának felmérésére. Minden mintát a HMW PAH prevalenciája jellemez, ellentétben a legtöbb közölt vizsgálattal. Ezenkívül a laboratóriumi körülmények között végzett kísérleti kezelés összehasonlítható referenciaként szolgált a helyszínen gyűjtött mintákkal

V. zöldikét (*Chlorophtum comosum* L.) beltéri bioakkumulációs vizsgálatokra való alkalmazhatóságát vizsgáltam meg. Szignifikáns különbségek találtam a főzési gyakorlatok és az akkumulált mennyiségek között. Azokban a háztartásokban, ahol nagyobb volt a felhasznált napraforgóolaj mennyisége, a HMW PAH-ok relatív aránya lényegesen magasabb volt, mint a disznózsírt és vajat használókban. A rákkeltő B(a)p-t kimutatható mennyiségben mértdm, ahol a sütést a leggyakrabban használt főzési gyakorlatként alkalmazták.

A C. *comosum* mint tesztnövény képes volt LMW és HMW PAH-ok felhalmozására is, így érzékeny biomonitornak bizonyult a beltéri PAH szintek mérésére. Biomonitorként való használatát tovább növeli ennek a dísznövénynek a népszerűsége és könnyen termeszthető jellege.

A kapott eredmények alapján megállapítható, hogy a protokoll használatával fontos információk gyűjthetők az egyes vizsgált zöldségek akkumulációs mintázatáról, és egyértelmű dózis-hatás összefüggések rajzolhatók ki. A szennyezett zöldségfogyasztás tényleges kockázatának felméréséhez azonban terepvizsgálatokra van szükség.

Zusammenfassung

Als Folge von Zunahme der Verstädterung in Europa und weltweit und der immer größeren Anzahl der Kraftfahrzeuge ist die Luftqualität in Großstädten in erhöhtem Maß verschlechtert worden wegen der verkehrsbedingten Aerosolpartikeln (schwebende Staubpartikeln). Diese schwebende Staubpartikel mit einer Größe von kleiner als 10 µm bzw. 2,5 µm (PM10 bzw. PM2,5), sowie die Luftverschmutzungskomponente auf deren Oberfläche stellen einen potenziellen Risikofaktor für die Pflanzen dar. Für die Untersuchung der Luftqualität können neben der Datensammlung durch Instrumente auch durch sogenannte passive Biomonitoring verwendet werden, d.h. das Ausmaß der Schadstoffbelastung kann anhand der in den Pflanzen akkumulierten Schadstoffe beurteilt werden.

Als Ziel meiner Forschung galt die Untersuchung der Bioakkumulation von Luftverschmutzungskomponenten unter kontrollierten Laborbedingungen bei Anwendung von Normenprotokoll. Meine Forschungsarbeit beinhaltet die Untersuchung der Verwendbarkeit der OECD Norm Nr. 227 für die Einschätzung der Bioakkumulation weiterhin die Feststellung der faktischen Bioakkumulationskapazität von ausgewählten Pflanzensorten. Dementsprechend habe ich folgende Untersuchungen durchgeführt:

I. Bei Laborbedingungen habe ich die Akkumulation von PAH-Verbindungen aus Aerosol im Gartensalat (Lactuca sattiva L.) untersucht. Insgesamt haben wir 19 PAH-Komponente in Salatproben gefunden, von denen 16 Komponente gelten als Prioritäten in den US EPA, von denen stellt die Dibenzo(a,h)anthracen die einzige Ausnahme dar, welches in keiner der Pflanzenproben detektiert werden konnte.

II. Das Bioakkumulationspotenzial von 8 verschiedenen Gemüsesorten habe ich miteinander verglichen bei kontrollierten Laborbedingungen. In dem Akkumulationspotenzial der verschiedenen Pflanzensorten konnte ich auffallende Unterschiede feststellen, was anhand der blättermorphologischen Parameter nur teilweise geklärt werden kann. Wahrscheinlich sind die akkumulierten PAH-Werte auch der verschiedenen Wachstumsrate zuzuschreiben. Das Akkumulationsmuster in den verschiedenen Gemüsesorten wies eine gewisse Ähnlichkeit auf, in jeder Pflanzensorte überwiegen die PAH-Komponente mit niedrigerem Molekulargewicht (LMW).

III. In kleinen und mittelgroßen Ortschaften im Komitat Veszprem habe ich durch Bioakkumulationsuntersuchungen das Ausmaß der Luftverschmutzung anhand von in Blumentöpfen gezüchteten Testsalatpflanzen festgestellt. Die in den Salatpflanzen akkumulierten PAH-Konzentrationen bewegten sich in einem breiten Bereich. Aufgrund der Ergebnisse ergeben sich als Hauptquelle der PAH-Emission die Verbrennung von Biomasse, die Haushaltsheizung und die Verbrennung von landwirtschaftlichen Abfällen in den Gärten. Dazu tragen auch die verkehrsbedingten Emissionen bei. Die Tatsache, daß sich der höchste Verschmutzungsgrad sowohl in den Salatpflanzen wie auch im Boden der Harskut Natura 2000 ergab, läßt unser Aufmerksamkeit darauf lenken, wie wichtig die Analyse der verschiedenen Verseuchungsquellen ist.

IV. In den Proben von Spitzwegerich (*Plantago lanceolata* L.) aus verschiedenen Probenentnahmestellen wie auch von im Labor gezüchteten Pflanzen habe ich die akkumulierten PAH-Werte festgestellt und die möglichen Verseuchungsquellen bestimmt. Aufgrund dieser Untersuchungen konnte bewiesen werden, daß die P. lanceolata kann als zuverlässiges passives Monitor für die Verteilungsanalyse der PAH-Verseuchung dienen. Die Prävalenz von HMW PAHs galt in allen Proben in Kontrast zu den meisten veröffentlichten Untersuchungen. Darüber hinaus konnte die Versuchsbehandlung in Laborbedingungen als Vergleichsreferenz für die an Ort und Stelle genommenen Proben dienen.

V. Für die Möglichkeit der innenräumlichen Untersuchungen von Bioakkumulation habe ich die Grünlilie (*Chlorophytum comosum*) verwendet. Signifikante Unterschiede konnte zwischen den verwendeten Kochpraxen und den akkumulierten Mengen festgestellt werden. Bei Haushalten, wo mehrheitlich Sonnenblumenöl benutzt wurde, die relativen HMW PAH-Werte lagen wesentlich höher als bei denen, die Schweineschmalz oder Butter verwendeten. Die C. *comosum* als Testpflanze hat sowohl LMW- als auch HMW-PAH-Komponente akkumuliert und so erwies sie sich als sensitives Biomonitor für die Bestimmung von innenräumlichen PAH-Werten. Die Verwendung dieser Zierpflanze als Biomonitor wird durch ihre Popularität und einfache Züchtung verbreitet.

Aufgrund der erzielten Ergebnisse kann festgestellt werden, daß durch die Anwendung des Protokolls wesentliche Informationen über das Akkumulationsmuster von den untersuchten Gemüsesorten gesammelt und eindeutige Zusammenhänge zwischen Dosis und Wirkung dargestellt werden können. Für die Einschätzung der tatsächlichen Gefährdung durch verseuchten Gemüseverbrauch sind aber weitere Felduntersuchungen nötig.

Хураангуй

Европ болон дэлхийн дахинд хотжилт нэмэгдэж, тээврийн хэрэгслийн тоо нэмэгдэхийн хирээр томоохон хотуудын агаарын чанар замын хөдөлгөөнөөс үүссэн аэрозолын тоосонцороос болж улам дордож байна. (тоосонцор - PM) Эдгээр агаарын тоосонцор нь 10 макрометр болон 2,5 макрометрээс бага хэмжээтэй ба түүнчлэн эдгээрээр гадаргуу дээр нь хуримтлагдсан бүрэлдэхүүн хэсгүүд нь ургамалд эрсдэл учруулдаг. Бохирдсон ургамлын бохирдуулагчийг хэмжихийн тулд өгөгдөл цуглуулснаар агаарын чанарыг хэмжиж, ангилах боломжтой болдог.

Миний судалгааны зорилго бол лабораторын нөхцөлд агаарын бохирдлын бүрэлдхүүний био хуримтлалыг стандартын дагуу судлах байсан. Би энэхүү судалгаагаараа сонгосон ургамлын бодит био хуримтлалын хэмжээг тодорхойлж, био хуримтлалыг ӨИСД 227 (Эдийн засгийн хамтын ажиллага болон хөгжүүлэх байгууллага) стандартын дагуу үнэлж, туршилт хийсэн. Үүнчлэн, дараах туршилтуудыг хийсэн.

I. Лабораторын нөхцөлд Шанцай (*Lactuca sativa* L.) ургамал дээр ПАХ (PAH) (Полициклик үнэрт устөрөгчид)-ын хуримтлалыг судалсан. Шанцайн ургамлын дээжинд нийт 19 ПАХ (PAH) илэрсэн, үүнээс 16 нь АНУ-ын ЕПА (US EPA) байсан. ЕПА (US EPA)-ны дагуу дибензо (a, h) антрацен илэрхийг таамаглаж байсан ч бидний дээжинд илрээгүй.
II. Био хуримтлалыг 8 өөр ургамал дээр харьцуулан лабораторын нөхцөлд судалсан. Ургамлуудад гайхалтай өөр өөр хуримтлалын үр дүн үзүүлсэн ба тэдгээрийг навчны морфологийн параметр үзүүлэлтээр тус бүр тайлбарлья. Магадгүй ургамлын өсөлтийн өөр өөр хэмжээнд ПАХ (PAH)-ын хуримтлалын концентраци нөлөөлсөн. Ургамлуудын хуримтлалын дээжүүдийн хувьд бага жинтэй молекул (LMW)-т ПАХ (PAH) нэгдлүүд давамгайлж, зарим ижил төстэй байдлыг харуулсан.
III. Шанзайны ургамал (*Lactuca sativa* L.)-аа жижиг болон дунд хэмжээтэй саванд үрслүүлж, агаарын бохирдлын түвшинг харуулах туршилт болох био хуримтлалын туршилтаа Веспрем хотод хийсэн. Шанцайны ургамал дахь хуримтлагдсан ПАХ (PAH) концентраци нь өргөн хүрээний хэлбэлзэлтэй байв. Биомассаар шатаах нь ПАХ (PAH) үүсгэх үндсэн үүсвэр болох нь судалгааны үр дүнгээс харагдсан ба эдгээрт өрхийн дулаан халаалт, хөдөө аж ахуйн хог хаягдлыг

ил шатаах зэрэг орохоос гадна тээврийн хэрэгслээс үүсэх бохирдол ч мөн орно. Hárskút Natura 2000 бүсэд шанцай ургамал болон хөрсний дээж хоёулаа их хэмжээний бохирдолтой байсан нь бохирдлын эх үүсвэрийг тус тусад нь шинжлэхийн ач холбогдолтой болохыг бидэнд илтгэж байна.

IV. Юлдэн ургамалд (*Plantago lanceolate* L.) хуримтлагдсан ПАХ (PAH)-ийн хэмжээг өөр өөр газруудаас цуглуулсан дээжүүдийг харьцуулан, лабораторын нөхцөлд туршилт хийсэн. Боломжит эх сурвалжийг уламжлалт эх сурвалжийг тодорхойлох аргыг ашиглан тодорхойлсон. Туршилтаар Юлдэн ургамал (*Plantago lanceolate* L.) нь ПАХ (PAH)-ийн бохирдлын тархалтын хэв маягийг үнэлэх найдваргүй, идэвхигүй монитор болох нь батлагдсан. Бүх дээжүүд их жинтэй молекул (HMW)-т ПАХ (PAH)-ийн тархалтаар тодорхойлогдож байсан нь ихэнх судалгаанд тэмдэгдсэн байна. Нэмж дурдахад, лабортарийн нөхцөлд хийсэн туршилт маань судалгааны талбараас цуглуулсан дээжтэй харьцуулах боломж бүрдсэн.

V. Аалзан ургамал (*Chlorophtum comosum* L.)-ыг тасалгааны био хуримтлалын туршилтад хэрэглэсэн. Би хоол хийх явц болон ПАХ (PAH)-ийн хуримтлалын хэмжээний хооронд мэдэгдэхүйц ялгаа буйг олж мэдсэн. Гэрийн нөхцөлд наранцэцгийн тосны хэрэглээ өндөр байх нь их жинтэй молекулт (HMW) ПАХ (PAH)-ийн хэмжээ мөн өндөр байгаа нь гахайн өөх, цөцгийн тос хэрэглэдэг өрхүүдээс давамгай байв. Гэрийн нөхцөлд жигнэмэг хийх явцад хорт хавдар үүсгэгчийг илрүүлж болох хэмжээнд хэмжсэн.

Протоколын дагуу гарсан үр дүнгээс харахад ургамал тус бүрийн хуримтлалын хэв шинж ба цэвэр тунгийн нөлөөний харилцан хамаарал чухал мэдээлэл өгч байна. Гэсэн хэдий ч, судалгаагаар бохирдсон хүнсний ногоо хэрэглэх бодит эрсдлийг үнэлэх хэрэгцээ байна.

1. Introduction

1. 1. General introduction

Since the end of the industrial revolution the rapid rate of industrialization and urbanization has caused the exponential growth of anthropogenic activitites. These activities had serious negative effect on the environment. With the increase of industrial activities the emission of various pollutants into the environment also increases. In the environment anthropogenic pollutants contaminate every spare of the environment. One of the spheres most affected by human pollution is the atmosphere.

Pollution in the atmosphere is also referred as air pollution. Air pollution has detrimental effects not just on the public health, but on the whole planet. The effects of air pollution can be divided into global and local effects based on the scale of the pollution. Global effects include global warming and climate change due to the emission of anthropogenic greenhouse gases, and the depletion of the ozone layer on the southern hemisphere. This depletion might be caused by because the emission of chlorofluorocarbons.

According to the studies of World Health Organization (WHO) air pollution is the biggest environmental threat to human lives because every year approximately seven million people's death can be associated with the breathing of insufficient quality of air (WHO 2021). Also, according to the WHO the air that nine out of ten people breath exceeds the WHO's guideline limits for pollutants (WHO 2016). In the European Union the Directive 2008/50/EC of the European Parliament and of the Council sets limit values for indoor air quality to regulate the emission of harmful air pollutants (EU. Directive 2008/50/EC).

Air pollution in settlements has become a steadily growing problem. Pollution from traffic, especially emission of diesel-powered vehicles, poses a serious health concern. In Hungary the average age of the cars was 14.7 years in 2020, diesel was used by the 18.5% of passenger cars as fuel but 2,230,859,742 liter of diesel were sold (Közlekedésbiztonság, 2023). Particles emitted from these sources carry a wide range of potentially toxic chemicals, like polycyclic aromatic hydrocarbons (PAHs) witch are one of the most important groups of concern.

1. 2. Particulate matter

Atmospheric particulate matter (PM) is a group which contains solid and liquid particles which are suspended in air. (Jaenicke, 2001). They can be also referred as atmospheric aerosol, or airborne aerosol. In the environment the PM can be evaluated by their mass concentration per unit of air, or the number of particles per unit volume of air. (mg or ng m^{-3} or n cm^{-3}).

Particulate matter generally shows high variability in terms of physical and chemical attributes. Their source in the environment can be either natural or anthropogenic. Depending on their sources, aerosol physical properties like size, mass, density, surface area will vary. PM chemical composition also shows high variability based on its origin.

Atmospheric particulate matter can be characterized by it residence time which is the time which it stays in the air. The residence time depends the PM physical and chemical properties.

Several types of PM classification exist. These classifications are mainly based on their origin or its generation process, or its physical or chemical properties.

Particulate matter can be classified by its formation mechanism into primary and secondary origin. Primary origin particulate matter is those which are directly emitted in to the environment as solid or liquid. Secondary origin aerosols are those which are formed in the atmosphere from gas either by chemical reaction or by condensation. (Warneck P., 2000)

Particulate matter can also be classified by the size of the particles as coarse, fine, and ultra fine. They can be divided in to groups based on their aerodynamic diameters. The course PM has bigger diameter than 10 um, they are also referred as PM10. The fine aerosol has smaller diameter then 2,5um and called PM2.5. The ultrafine particulate matter has a smaller size then 0,1um they are referenced in the literature as PM0.1 The adverse effect on the human health of the PM2.5 has been long studied. (e.g. Schwarz et al. 1996) The European Union, sets threshold for 10 um diameter (PM10) and for the 2.5um (PM2.5) particulars. (European Parliament 2008.)

Generali is agreed the with the decrease of the particle size the potential hazard effect becomes higher, because as the PM size decreases its relative surface area to mass increases, and with higher surface are it can bod relatively higher concentration of toxic compounds, and its easier for the small particulars to travel thru biological barriers. (Chen et. al. 2016)

Based on its origin atmospheric aerosols can be divided into two categories natural aerosols and anthropogenic aerosols. Natural aerosol is emitted from natura sources. Larger fractions of

natural sources come from desert and soil particles resuspension. Another large portion is from Vulcanic origin. The Vulcanic aerosol is made up mineral materials (Vulcanic ash) and seconder origin particles which are formed by the oxidation of sulphur oxides.

Another type of natural particulate matter is biogenic PM. They are also referred as bioaerosols. The are all produced by living organism mostly and made up of pollen, spores, and small microorganisms like age, bacteria, viruses. Most of the biogenic PM are primary aerosols, and their size falls in the course particles witch the only exceptions of viruses and small bacteria (Fennelly, 2020)

Anthropogenic aerosols are emitted in the atmosphere by the involvement of human activities. They take up a significant portion of the particulate matter. Anthropogenic PM sources are local and usually located in urban and industrial areas. Usually they propose a local effect, near the source of the emission. In urban areas the source of anthropogenic particles are associated with traffic and residential heating . Another form of anthropogenic emission of particles is fugitive emissions, which consist those resuspended powdery materials that enter the atmosphere doe to handling, like mining, constructional works, ore agricultural activities.

According to numerus studies the pollution from anthropogenic aerosols is one of the most major environmental concerns in Europe. (Vicente et al. 2018) Particulate matter with and elemental carbon core have an ability to adsorb a large number of potential toxic compounds. One of these compounds are polyaromatic hydrocarbons (PAHs) (Health Effects institute 2002.)

1. 3. Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are unsaturated hydrocarbons comprised in cyclic manner of carbon and hydrogen molecules. They contain at least two infused aromatic rings. PAHs as a group is made of more than 200 congers generally can be found in complex mixtures not in single compounds. PAHs in the environment are recognized as persistent organic pollutants (POPs) by the Stockholm Convention (Stockholm Convention 2011). Most of the PAHs are chemically inert. They cannot burn easily, while PAHs can be degraded under ultra violate light. PAHs can also react in air with tropospheric hydroxyl radicals, ozone, sulfuric acid, and nitrogen oxides. Table 1 gives basic data for 16 PAHs classified as hazardous by the Environmental Protection Agency of the United States.

Table 1. The physicochemical properties of EPA 16 PAHs.

Compound name	Chemical formula	Molecular weight (g mol^{-1})	Meling point (°C)	Boiling point (°C)
Naphthalene	$C_{10}H_8$	128.2	80	218
Acenaphthylene	$C_{12}H_8$	152.2	92-93	265-275
Acenaphthene	$C_{12}H_{10}$	154.2	203	279
Fluorene	$C_{13}H_{10}$	166.2	116-117	295
Phenanthrene	$C_{14}H_{10}$	178.2	100	340
Anthracene	$C_{14}H_{10}$	178.2	218	340-342
Fluoranthene	$C_{16}H_{10}$	202.3	111	375
Pyrene	$C_{16}H_{10}$	202.3	156	393-404
Benzo[a]anthracene	$C_{12}H_{12}$	228.3	158	438
Chrysene	$C_{18}H_{12}$	228.3	254	448
Benzo[b]fluoranthene	$C_{20}H_{12}$	252.3	168	481
Benzo[k]fluoranthene	$C_{20}H_{12}$	252.3	216	480
Benzo[e]pyrene	$C_{20}H_{12}$	252.3	177	492
Benzo[a]pyrene	$C_{20}H_{12}$	252.3	179	495
Indeno[1,2,3-cd]pyrene	$C_{22}H_{12}$	276.3	164	530
Dibenzo[a,h]anthracene	$C_{22}H_{14}$	278.3	262	535
Benzo[g,h,i]perylene	$C_{22}H_{12}$	276.3	273	550

PAHs are generated during incomplete combustion of organic material. The crucial factors in the role of the process also includes high temperature, and the lack of proper amount of oxygen for the complete burning. The process in which PAHs are formed generally results in a complex mixture of different PAH congeners (Sampiano et al. 2021). PAHs are characterized by low water solubility, low vapor pressure, high melting, and boiling points depending on their structures (Lee and Vu 2010). With the increase of their molecular weight, the water solubility tends to decrease, and the lipophilicity tends to increase (Okere and Semple 2012). They can be classified according to the number of aromatic rings as light (low molecular weight) (LMW) PAHs and as high (high molecular weight) (HMW) PAHs. Low molecular weight PAHs are comprised of 2 and 3 ring PAHs while high molecular weight PAHs contain 4 to 6 rings (Purcaro et al. 2013). Depending on their volatility and molecular weight, PAHs are emitted as gas phase (LMW PAHs) or bonded to particles (HMW PAHs) (Lee and Vu 2010). Particulate matter with and elemental carbon core have an ability to adsorb a large number of potential toxic compounds. One of these compounds are polyaromatic hydrocarbons (PAHs) (Health Effects Institute 2002).

Particulate matter is in general considered as a serious environmental hazard. Atmospheric particulate matter (PM) is a group which contains solid and liquid particles which are suspended in air (Mészáros 1999). They can also be referred as atmospheric aerosol, or airborne aerosol.

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It is generally agreed that with the decrease of the particle size the potential hazard effect becomes higher, because as the PM size decreases its relative surface area to mass increases, and with higher surface area it can bind relatively higher concentration of toxic compounds, and its easier for the small particles to travel through biological barriers (Chen et. al. 2016).

PAH sources

Classification of PAH emissions sources

In the air PAHs can occur in two different states: in gas phase or bonded to particles. PAHs with higher volatility with two or three rings have higher vapor pressure and can occur in the gaseous phase while PAHs with higher boiling point, lower volatility and lower vapor pressure (mainly four, five and six ring PAHs) have a tendency to bind to particles (Mukhopadhyay et al. 2020).

PAHs in the environment can accumulate in the water, air, and soil, and can enter into the food chain (Zelinkova and Wenzl 2015, Abdel-Shafy and Mansour 2016). Environmental PAHs distribution and toxicity are largely affected by their emission sources. PAH emission sources can be characterized based on several attributes. Based on the movement of the emission source it can be divided into stationary sources, and mobile sources. Based on the origin of the emission PAH pollutions can be categorized mainly as anthropogenic emission sources and natural emissions (Mojiri et al. 2019). Based on its sources anthropogenic PAHs can be further divided into four groups i.e., industrial, mobile, domestic, and agricultural (Ravindra et al. 2008).

Natural sources

PAHs can form naturally from any natural occurring fires caused by lightning like forest fires, bush fires. Natural emissions also include volcanic eruptions and decaying organic matter, and biogenic PAHs which are produced by living organisms like certain plant and bacteria. The amount of PAH production is depending on meteorological conditions like wind, humidity, temperature, and the characteristics of combustible material, like wood type, wood moisture content, and seasonal wood. These natural PAH sources are less important and their amount are much smaller than anthropogenic sources (Patel et al., 2020). Anthropogenic PAH sources are the most dominant source of environmental PAHs than natural PAHs.

Anthropogenic sources

In urban areas, vehicular traffic and biomass burning have been reported as the main contributors to PM emissions (Cipoli et al. 2023). Although exhaust emissions have shown a diminishing tendency in Europe due to more stringent regulations coupled with technological improvements such as the use of diesel particulate filters (DPF), limit values are still often exceeded in cities. Naturally, people living in areas most impacted by high traffic will show the

most severe symptoms of disturbance. As such, traffic-related pollution has been in the focus of both researchers and environmental decision-makers.

In urban environments, diesel exhaust has been shown as one of the major sources of fine and ultra-fine particulate matter (PM2.5 and PM0.1). According to estimations, in large cities up to 50 % of PM2.5 mass concentrations can be linked to traffic-related emission sources (Chen et al. 2001). This higher PM concentrations were contributed from the exhaust (tailpipe) and non-exhaust (e.g., tyre and brake wear, re-suspension of dust) emissions of vehicles (Pant and Harrison, 2013; Kumar et al., 2021) in traffic-related microenvironments. Diesel engine exhaust has been claimed as carcinogenic to humans (Group 1) by the International Agency for Research on Cancer (IARC) (IARC 2012). In comparison with gasoline vehicles, diesel vehicles provide more significant emission sources of urban air PAHs (Barakat 2002).

In order to control PM emission of vehicles, environmental standards have been introduced in the European Union for passenger cars and for heavy-duty vehicles (Table 2.).

Table 2. Environmental standards for diesel passenger cars and heavy duty vehicles

Standard	Introduced for diesel passenger cars	PM10 for diesel passenger cars [mg km $^{-1}$]	Introduced for diesel heavy duty vehicles	PM10 for diesel heavy duty vehicles [mg kWh^{-1}]
EURO 1	1993	140	1991	360
EURO 2	1997	80	1996	150
EURO 3	2001	50	2001	100
EURO 4	2006	25	2005	20
EURO 5	2011	5	2008	20
EURO 6	2015	5	2012	10

When assessing eco- and genotoxicity of PM10 emission of buses belonging to different emission standard categories, it was found that both eco- and genotoxic potential of the emissions were highly depending on the standard (Kováts et al. 2013). Results showed that ecotoxicities of Euro0–Euro1 engine emissions fall close to each other, in the range of 1.24–0.96 μg ml^{-1}, while emission of Euro4 vehicles proved to be non-toxic. Genotoxic potential estimated by the colorimetric SOSChromotest™ showed correlation with the ecotoxic potential.

A similar study was conducted on emissions of passenger cars belonging to different emission standard categories. The study found a good correlation between ecotoxicity of the emissions

and PAH content. However, environmental hazard of the selected cars was strongly depending on the age and odometer reading in addition to Euro standard (Ács et al. 2013).

A wide range of studies have addressed seasonal differences in PAH concentrations and have found considerable seasonal variations, with highest values in cold seasons (Morville et al. 2011). It can be attributed mostly to higher contribution of combustion-related emissions but other reasons also apply, such as lower wind speed, lower atmospheric mixing heights, or lower rate of photochemical oxidation by solar radiation (Teixeira et al. 2012). The highest seasonal difference for particle-phase PAHs was observed when seasonal pattern of both vapor- and fine-particle–phase PAHs were compared Eiguren-Fernandez et al. (2004). Hong et al. (2007), however, reported that the main sources of particle bound PAHs were mainly vehicular exhaust, regardless of season. Our previous study also revealed higher environmental impact of particulates generated in colder periods, showing that biomass burning could be the most important source for spring and autumn samples (Kováts et al. 2020).

In addition to seasonal patterns, diurnal patterns in PM and related PAHs emissions are also often discussed. Cipoli et al. (2023) for example found differences in nighttime-daytime emission rates and patterns, as at nighttime emissions were dominated by residential biomass combustion while at daytime vehicle traffic was the dominant contributor.

Domestic PAH sources are major contributors to the total emission of anthropogenic PAH in the environment. The term ‘domestic emissions’ refers to household activities like cooking and residential heating (Srogi et al. 2007; Ravindra et al. 2008; Patel et al. 2020). The burning and pyrolysis of oil, coal, gas, wood, garbage, and other organic materials are the main domestic sources. There is a large geographic variation in domestic emission which is due to the differences in climate pattern and domestic heating systems. Major indoor PAH sources are for emissions from cooking and residential heating.

The emission of PAHs from domestic sources is major concern of public health because they are significant sources of indoor emissions (Ravindra et al. 2006). Diffusion of PAHs from outdoor sources is also an important factor affecting indoor air quality. A study assessed the distribution of PAHs in indoor nonsmoking house dust and estimated that 4 ring PAHs predominated the samples accounting 40-53% of total PAHs followed by 3 ring PAHs 29-40% of total PAHs.

Another dominant source of indoor PAHs originates from cigarette smoke. According to several studies indoor air in smoking residence shows higher PAHs concentrations than non-smoking residences (Hoh et al. 2012). It was estimated that cooking can be responsible for 32.8% of total indoor PAHs (Zhu et al. 2009). According to a report of the World Health Organization (WHO) in China, India and South East Asia over 75% and in South America and Africa 50%-75% of population uses solid fuels like wood for cooking.

Coal stoves are still a widely used for cooking and heating in China's Northern rural parts (Liu et al. 2008). In this study the total emission factors (EFs) of the measured 15 PAHs ranged from 52.8 to 1434.8 mg kg^{-1}. The results depended on the dryness and composition of coal that was uses as cooking and heating fuel. Another study found major decrease in emission factors of PAHs from residents with coal stoves which used honeycomb coal briquettes compared to regular coal (Chen et al. 2004). An early study compared emission factors from wood fuel, charcoals, coal briquette in a resident stow and found that wood fuel burning released the highest PAHs levels and charcoal released the lowest amount of PAHs (Oanh et al. 1999). In the study they found 18 PAHs from which 11 was genotoxic. The emission based on the fuel weight showed the similar emission factors of wood fuel and coal briquette 110 mg kg^{-1}, although the concentration of genotoxic PAHs was twice as high in wood fuel burning compared to charcoal burning.

Industrial PAHs are produced by incomplete combustion during various industrial production lines. Industrial processes, where fuels such as gas oil coal and wood are burned produce excessive amount of PAHs. Another major source of PAHs emitted during various processing of raw materials like aluminium processing coke production. PAH compounds are not synthetized directly as a chemical product. Some PAHs are used in various industrial productions as interceders in pharmaceutical industries or as dyes in paint industries, plastic industries (Kaminski et al. 2008.) Acenaphthene, Anthracene, Fluorene, Phenanthrene Pyrene, are used during production of pigments, dyes, pesticides and wood preservations. Elevated concentrations of PAHs have been found in bitumen and asphalt production plants, smoke houses, coking plants, and near oil refineries.

Other major sources from the industrial processes include incineration of wastes, cement production, rubber tire manufacturing industries, power production in power plants (Patel et al. 2020; Amirdivandi et al. 2019; Srogi et al. 2007). PAHs are generally found in coal-tar

production plants, petroleum products or coal, or where wood, or other plant materials are burned.

Agricultural PAHs sources mainly come from open biomass burning, and biomass burning, when the burning is lacking sufficient amount of oxygen (Ravindra et al. 2008). In rural areas the high PAH pollution emission is mainly caused by agricultural and domestic sources, where the pollution in urban area mainly comes from industrial, mobile, and domestic sources (Patel et al. 2020).

In addition to 'traditional' sources such as biomass burning, the illegal practice of burning household waste has become a worldwide source of PAH emissions. It is a common problem detected both in developed and developing regions of the world. Lemieux (1998) for example analysed the practice in the USA commonly referred to as barrel burning, that is, locals burn their waste in the open in barrels. According to the study, the motivations are in most cases simple convenience, or cost avoidance. Open burning is generally common in areas that have volume-rate disposal policies, implying that households have to pay for disposal proportionally to the amount of waste generated (Park et al. 2013).

Depending on the composition of waste, significant amount of PAHs can be generated. Lemieux (1998) open burned domestic waste including different plastic materials under experimental conditions. Plastic amounted to app. 7.5% of the total waste in a non-recycler and 15% in an avid recycler household. The study compared PAH emissions between open burning of household waste and controlled combustion of municipal waste in a municipal waste combustor (MWC). While PAH emissions from the MWC were 16.58 mg kg^{-1} waste burned, these values were as high as 66,035.65 mg kg^{-1} waste burned (!). Kováts et al. (2022) selectively burned different waste types under controlled conditions and found high PAH emission in case of some plastics such as Polypropylene, Polystyrene and Polyurethane. Genotoxicity was also assessed using the SOSChromotest and SOSIF values (indicators of genotoxicity) showed good correlation with several individual PAHs. The study concluded that waste emissions rich in PAHs pose high risk of mutagenicity either burning indoor or outdoor.

The concentration of PAHs shows seasonal patterns. The highest concentration is the highest in winter than followed by spring, autumn, and summer. The high concentration in winter and spring is caused by the incomplete combustion of the burning of fossil fuels, caused by higher

amount of indoor heating, and lower levels of photodegradation do to the lack of light, and the poor diffusion in the air caused by calm winds and low temperatures (Patel et al. 2020).

PAH sources can also be categorized by the origin of production into three categories i.e., pyrogenic, petrogenic, and biogenic (Mojiri et al. 2019).

Pyrogenic PAHs are formed during incomplete combustion of organic matter at high temperatures (350-1,200°C) under insufficient amount of oxygen (Patel et al. 2020). Major sources of pyrogenic PAHs include burning of fossil fuel (coal, oil, petroleum) and biomass burning. Other pyrogenic PAHs sources include pyrolytic synthesis processes like thermal braking in petroleum production where long chain hydrocarbons are broken down into lighter hydrocarbons, coal tar distillation, and coke production (Patel et al. 2020). Petrogenic PAHs originate form crude oil and its by-products and enter the environment due to mining, transport, storage leakage (Patel et al. 2020). Biogenic PAHs are produced by living organisms like microorganisms and phytoplankton (Mojiri et al. 2019).

PAH Source appointment

Certain PAH compounds form with higher probability in either pyrogenic or petrogenic situations. In pyrogenic sources HMW PAHs are predominant while in petrogenic sources the LMW PAHs are more dominant (Marris et al. 2020).

The ratio of certain PAHs can give some indication about the source(s) of the emission, the technique and actual ratios were first described by Yunker et al. (2002). Since then, some modifications have been introduced (e.g. Tobiszewski et al. 2012), however, the original approach of Yunker et al. is being used hereinafter.

The method is based on calculating the ratio of traditionally established PAH pairs such as Flt/(Flt+Pyr), BaA/(BaA+Cry), and Ind/(Ind+BghiP). The ratio of Flt/(Flt+Pyr) below 0.4 likely implies petroleum input, between 0.4 and 0.5 petroleum combustion while ratio above 0.5 indicates grass/wood and coal combustion. BaA/(BaA+Cry) ratio below 0.2 indicates petroleum, between 0.2 and 0.35 either petroleum or combustion implying mixed source and ratio above 0.35 indicates fossil fuel and vegetation combustion. Ind/(Ind+BghiP) ratio higher

than 0.5 likely implies combustion of grass, wood and coal between 0.2 and 0.5 petroleum combustion and ratio lower than 0.2 indicates petroleum source.

These ratios have been also used to identify possible PAH sources in plant samples. Deelaman et al. (2023) reported that open burning of agricultural wastes had a significant contribution to accumulated PAHs in rice grains in Thailand and Laos. Zhang et al. (2018) analysed PAH concentrations in cabbage collected near a large coking manufacturer in Shanxi Province, Northern China, and demonstrated coal combustion as the main input source. Most studies, however, use these diagnostic ratios to identify pollution sources and potential risk in agricultural soils (e.g. Chai et al. 2017).

PAHs Health effects

Several polyaromatic hydrocarbons have proven to be mutagenic and carcinogenic. There is a group of PAHs that are notorious for their adverse effect which are the so called Car-PAHs, which includes benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene (B(a)P), dibenzo(a,h)anthracene, indeno(1,2,3-cd)pyrene, benzo(g,h,i)perylene, and chrysene (Srogi et al. 2007). In the 1970s the US Environmental Protection Agency (EPA) identified 16 priority PAHs (EPA-PAH) which are often found in environmental monitoring samples: acenaphthene, acenaphthylene, anthracene, fluoranthene, fluorene, naphthalene, phenanthrene, pyrene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[g,h,i]perylene, benzo[a]pyrene, chrysene, dibenzo[a,h]anthracene, and indeno[1,2,3-cd]pyrene. In the European Union, the European Scientific Committee on Food (SCF) recommended the monitoring of 15 PAHs: benzo[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[g,h,i]perylene, benzo[a]pyrene, chrysene, cyclopenta[cd]pyrene, dibenzo[a,h]anthracene, dibenzo[a,e]pyrene, dibenzo[a,h]pyrene, dibenzo[a,i]pyrene, dibenzo[a,l]pyrene, indeno[1,2,3-cd]pyrene, and 5-methylchrysene which have been proven mutagenic/genotoxic 64 in animal experiments (EU priority PAHs) (reviewed by Zelinkova and Wenzl 2015).

In order to quantify possible human health hazards, toxicity equivalency factors (TEFs) were already proposed by Nisbet and LaGoy (1992). B(a)p serves as a reference chemical and is assigned a value of 1 while TEF values of other PAHs are calculated based on their carcinogenic level in comparison to that of BaP. In case of complex samples, the carcinogenic potency of

total PAHs is obtained by summing the toxic benzo[a]pyrene equivalent (BaPeq) concentrations of each PAHs (Soltani et al. 2015).

1. 4. Bioaccumulation

The risk of bioaccumulation of polyaromatic hydrocarbons (PAHs) in vegetables was already reported in early studies (e.g. Voutsa et al. 1996; Kipopoulou et al. 1999).

A wide range of studies have assessed bioaccumulation in different vegetable species as they might pose human health risk when consumed. Results may vary, however, Khillare et al. (2012) for example found higher accumulation of 16 high priority atmospheric PAHs in vegetables which were grown in the neighbourhood of coal-based thermal power plants than in samples from background locations. High accumulation was also reported by Abou-Arab et al. (2014). They investigated the concentration of PAHs in vegetable leaves collected from different regions in Cairo (Egypt) and detected high amounts of carcinogenic PAHs except pyrene. On the other hand, low accumulation has also been reported: Tusher et al. (2021) e.g. investigated the concentrations of PAHs in red amaranth and spinach samples from rooftop gardens in the urban and peri-urban areas of Bangladesh and found negligible accumulation. Similarly low accumulation rate was detected in the study of Amato-Lourenco et al. (2017) in vegetable samples (spinach and collard green) collected in urban gardens of Sao Paulo (Brazil).

Comparative studies are also available on the accumulation potential of different vegetables such as the study of Jia et al. (2018). They collected samples near industrial areas of Shanghai and found in vegetables that the total concentrations of 16 PAHs ranged from 65.7 to 458.0 μg kg^{-1}. In the leafy vegetables the highest accumulation was shown in spinach (*Spinacia oleracea* var.) (223.3-458.0 μg kg^{-1}), followed by Chinese cabbage (*Brassica rapa* var.), (206.0-348.1 μg kg^{-1}), Shanghai green cabbage (*Brassica chinensis* var.), (206.4-284.7 μg kg^{-1}), and finally lettuce (*Lactuca sativa* L.) (132.0-319.2 μg kg^{-1}).

Environmental impact of agricultural biomass burning was assessed by Chen et al. (2018). The study measured PAH accumulation in a wide range of vegetable samples including lettuces, cabbages, and Chinese cabbages taken in Chinese home gardens affected by straw burning. In lettuce samples, the concentration of total PAHs was in the range of 208.7 to 269.5 ng g^{-1}, in the range of 200.8 to 295.1 ng g^{-1} in Chinese cabbage and finally in the range of 114.5 to 175.1 ng g^{-1} in cabbage.

Brassicaceae species (e.g. *Brassica parachinensis*, *B. chinensis*, *B. juncea*) are widely used in bioaccumulation studies as they are generally considered good accumulators of PAHs (Franzaring 1997; Xiong et al. 2017; Zhang et al. 2018). In a study published by Mo et al. (2009) various *Brassica* species were collected from nine farms close to the Pearl River Delta in South China to evaluate the concentration of 16 PAHs in vegetable samples. The accumulated PAHs concentration were as follows (mean values are given): *Brassica parachinensis* (flowering Chinese cabbage) 438 μg kg^{-1}, *B. chinensis* (paitsai) 950 μg kg^{-1} and *B. juncea* (mustard) 1790 μg kg^{-1}.

Similar set of crop species was used in a case study of from Shunde (China) (Li et al (2008). PAH accumulation was measured in leafy vegetables including lettuce and Brassicaceae species such as cabbage, mustard and chinese flowering cabbage. The total concentration of accumulated PAHs was in the range of 128–1.258 μg kg^{-1} (not specified by species). In some studies extremely high amounts were found. In the study of Zhang et al. (2004), 16 EPA PAHs were analysed in vegetable samples from Minjiang River Estuary and the total PAH concentrations were between 8.600 to 111.000 μg kg^{-1} dry wt. in vegetable samples (not specified by species).

Bioaccumulation studies can also be used for assessing human health risk posed by consuming contaminated vegetables. These studies are utmostly important as diet is supposed to be the major source of human exposure to PAHs (Phillips 1999). Zhu et al. (2021) measured PAHs in edible parts of vegetables such as carrot and cabbage and their human health risks in Jinzhong (China) and concluded that dietary exposure probably implied high health risk. Interestingly, women exhibited slightly higher risks than man. In the above-mentioned study of Khillare et al. (2012), incremental lifetime cancer risk (ILCR) was calculated for vegetables grown in the vicinity of thermal power plants in Delhi (India) and actual risk appeared for lifetime ingestion exposure to PAHs.

In some cases, the magnitude of accumulation of toxic compounds can make consumption hazardous: Tesi et al. (2021) even concluded that vegetables consumed in some parts of Nigeria could be unsuitable for consumption based on their PAH, especially benzo[a]pyrene content. Wang et al. (2018a, b) also assessed human health risk in Beijing (China) based on 80 vegetable samples procured in local markets. However, the incremental lifetime cancer risk was below the acceptable risk level. In this study children were identified as the most sensitive group.

1. 5. Uptake

The accumulation of PAHs in plants in the environment can occur mainly from the polluted air or the polluted soil. The habitat of the plants can play a major role in the amount of contaminants which accumulate in plants.

Plants are exposed to both phases. Furthermore, PAHs from the atmosphere are easily transported to the soil, providing an additional exposure route for plants (Kulhánek et al. 2005).

Two well-known uptake mechanisms can be summarised as 1. from soil trough the root and transport to leaves, 2. from air with atmospheric deposition through the stomata (Kulhánek et al. 2005, Zhang et al. 2017a). The uptake of PAHs from gas-phase air and solid particles suspended in the air represents the main route by which organic compounds reach the above ground parts of plants (Kipopoulou et al. 1999; Tao et al. 2006a; De Salme et al. 2011a). PAHs in gaseous phase can diffuse through the wax and the cuticular membrane into the interior parts of the leaves and they can also enter directly through the stomata (Lehndorff and Schwark 2004). It is hypothesised that the transportation of PAHs from foliar deposition to the cuticular wax could be the primary pathway of leaf accumulation (Yang et al. 2017a). The uptake of lipophilic organic air pollutants like PAHs from the gas phase by plants is generally much more important than the particle deposition onto plant surfaces (Figure 1.) (Ortiz et al., 2023).

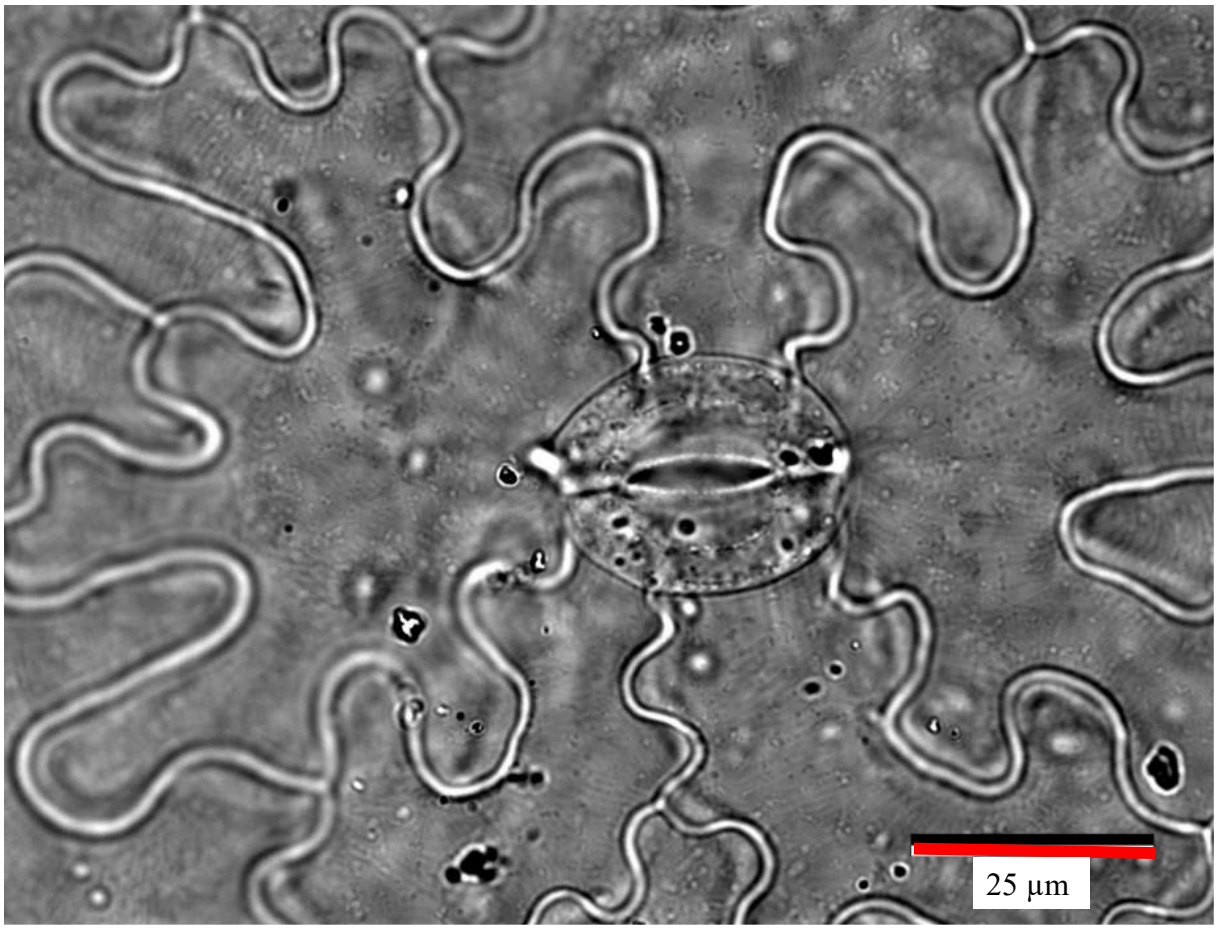

Figure 1. PAHs bonded to particles entered in to stomate by gas exchange and accumulated in plant leaf tissue (photo was taken by Zsófia Békéssy).

PAH concentrations in the aerial parts of plants are considerably higher than those found in the roots (Vácha et al. 2010), especially for plants with large leaf surface areas. The higher PAH concentration indicated that atmospheric deposition may strongly affect PAH concentrations in aerial plant parts (Edward 1983; Tao et al. 2004; Zhang et al. 2018). High PAH concentrations in internal leaf tissues can be attributed to the uptake of PAHs from atmospheric air by leaves and impaired translocation of those hydrophobic compounds, some of which are relatively persistent (Desalme et al. 2011a, b). Wei et al. (2021) examined the accumulation capacity of 11 plant species, they found evidence of combined pathways but higher PAH concentrations were determined in leaves than in other tissues for most of the investigated plants.

Based on the study of Kipopoulou et al. (1999) the adsorption from gas phase air could be the main source of the 2 and 3-ring volatile compounds (e.g. naphthalene, acenaphthene, fluorene, phenanthrene and anthracene), because these PAHs may also readily volatilise from polluted soil onto foliage.

The high molecular non-volatile compounds remain attached to particles and may be washed off again from the vegetation, high molecular compounds do not readily degrade and over time they may accumulate in the topsoil (Franzaring & van der Eerden, 2000).

The lower concentration in plant tissues of heavy PAHs with 5 and 6 rings were demonstrated by Li et al. (2017). The cuticle can modify the mobility of organic substances during transport. However, it shows size dependence, so an increase in the molecular weight results in a decrease in mobility. Oak leaves and pine needles for example showed higher accumulating capacity for light and medium molecular weight PAHs (Huang et al 2018).

In summer, the volatility of lighter PAHs from soils and vegetation could increase the concentrations in the above-ground parts of plants (Prevedouros et al. 2004; Tao et al. 2006b).

The investigation of the relationships between bioconcentration factors and the physicochemical properties of PAHs showed that solubility and the octanol–water partition coefficient are strongly correlated with the soil-to-root bioconcentration factors, while vapour pressure and the octanol–air partition coefficient were proved to be good predictors for the air-to-leaf accumulation of PAHs (Xiong et al. 2017).

Studies indicate that PAHs can be accumulated by plants from contaminated soils via their roots (Qamar et al. 2017; Zheng et al. 2014). However, the transport of PAHs from the roots to the above-ground parts of plants is relatively low (Gao & Zhu 2004; Kipopoulou et al. 1999; Wild et al. 2006; Wieczorek and Wieczorek 2007).

1. 6. Biomonitoring

Biomonitors by definition are organisms which can be used for the quantitative determination of environmental pollution (Markert et al. 2003). Biomonitoring or biological monitoring should be clearly differentiated from bioindication which according to Markert et al. (2003) gives qualitative information on the status of the environment.

Some authors, however, do not make such clear distinctions. Mukhopadhyay et al. (2020) give the following quasi-definition: ‘Accumulative bioindicators are organisms, groups of organisms or biological communities that help make the qualitative or quantitative assessment of ambient pollution due to the presence of contaminants and exhibit prominent morphological changes …’ Even worse, Rai (2016) gives the following and completely misleading statement:

‘Biomonitoring allows continuous observation of an area with the help of bioindicators’. For the sake of clarity, the approach of Markert et al. (2007) will be used hereinafter.

In practical terms, concentration of selected elements is measured from the sample organisms. Biomonitoring can be applied for assessing environmental status of different media such as soil, sediment, water or the air.

Biomonitors are often regarded as alternative to instrumental monitoring. Instruments are expensive and their use requires physical installation and energy supply (Augosto et al. 2013). This method is referred to as active monitoring or active sampling. Passive air samplers can be (1) artificial structures such as polyurethane foams or (2) living organisms called biomonitors. In general, passive samplers are relatively cheap and easy to handle. They can be used in remote areas as they do not need electricity supply (reviewed by Domínguez-Morueco et al. 2017).

Biomonitors allow to take numerous samples, due to their low cost. As such, high spatial resolution can be achieved. Also, studies requiring parallel measurements at numerous sites can be implemented (Lehndorff and Schwark 2004). In the European Union, Directive 2004/107/EC (2004) has proposed the use of biomonitoring approaches to map PAHs contamination (EC 2004).

Plants are exposed to both gaseous and particle-bound PAHs and they can reportedly accumulate both particle or vapour-phase PAHs. Lichens were the first to be used to assess air quality back in the 1860’s (Nylander, 1866). They are symbiotic association composed of algae (Cyanobacteria or Chlorophyceae) and fungi (Ascomycetes or Phycomycetes or Basidiomycetes). They show taxon dependent sensitivity to air pollutants called toxitolerance. Their sensitivity is partially due to the lack of defensive tissues, and they are exposed directly to air pollutants. In general, lichens have been the most widely applied in bioindication (Boonpeng et al. 2018) and studies reporting lichen biomonitoring results are still booming. In addition to PAHs, they effectively accumulate heavy metals, too. As they are epiphytic organisms, they can be easily transplanted to places which require attention but lack native lichen populations.

Similarly to lichens, mosses are extremely vulnerable to air pollution due to their morphology. Mosses have no roots therefore they are largely dependent upon wet and dry atmospheric depositions as sources of water and nutrient supplies. As mosses are often missing from highly impacted places, they are generally used in form of moss bags which are in fact transplanted

mosses. However, several studies have reported that mosses are effective in accumulating HMW PAHs which might be explained that (1) LMW PAHs degrade easily after entering the body of the moss and/or the dominant uptake mechanism in mosses is more likely the trapping of particles which typically bind HMW PAHs (Tretiach et al. 2011). Capozzi et al. (2017) conducted a comparative study in the area of Naples (Italy) and found that the deciduous tree *Robinia pseudoacacia* was able to accumulate both LMW and HMW PAHs, while in moss samples prevalently HMW PAHs were detected.

Lehndorff and Schwark (2009) used pine needles to follow changes in seasonal PAH-emission over an accumulation period of 4 years in the highly urbanized and industrialized Greater Cologne Conurbation (Germany). As samples were taken seasonally, the study could differentiate between summer and winter periods. Also, environmental behaviour of different PAHs could be analysed. As different pollution sources were typical in the area, the results could also be used for source appointment. Jeffrey pine trees (*Pinus jeffreyi*) were used in a similar study in Fresno, California to map the distribution of phenanthrene, which is an abundant PAH in the atmosphere (Noth et al. 2013). Using this widely distributed species, it was possible to conduct a systematic sampling based on the grid of 1-square mile United States Public Land Survey System blocks. In a comparative study of Klingberg et al. (2022) carried out in the urban area of Gothenburg, Sweden, black pine (*Pinus nigra*) showed much better accumulation potential than pin oak (*Quercus palustris*). Conifers have better capacity to capture larger amounts of particulate matter than broadleaved trees due to the morphology and number of leaves (Freer-Smith et al. 2005).

Quite many evergreen deciduous shrub or tree species have also been tested for their biomonitoring potential. Similarly to pines or spruces, they have thick epicuticular wax layer. In European context, taxa occurring in the Mediterranean region can be useful. Holm oak (*Quercus ilex*) was for example used to map the distribution of PAH contamination and to discriminate pollution sources in urban and periurban areas of Naples and Salerno in Campania region (Southern Italy) De Nicola et al. (2011). These evergreen (leaves overwintering) species can also be used to analyse seasonal patterns in PAH pollution. It should be noted, however, that use of herbaceous plants has been rather restricted (reviewed by Srogi 2007).

The choice of biomonitor species can be based on morphological and ecophysiological characteristics described earlier. Selecting crops can provide additional information: not only the status of the environment can be evaluated but even human health risk assessments can be

performed calculating the risk posed by consuming affected vegetables. Finally, in some cases local species can be used representing native, local flora, assessing impact on the actual recipient ecosystem (Sojinu et al. 2010).

Naturally, there are drawbacks in applying biomonitors, especially when studies of different authors need to be compared. De Nicola et al. (2016) argue that one of them is the lack of a standardized analytical methodology as different plant matrices require specific preparation. In general, experimental approaches are diverse making inter-study comparisons extremely difficult (Doucette et al. 2008). As such, several attempts have been made to standardise experimental protocols (e.g. Weber et al. 2018). The standardised ryegrass (*Lolium multiflorum*) method has been in use for monitoring PAH contamination in different areas such as e.g. Germany (Rodriguez et al. 2010), France (Crépineau-Ducoulombier et al. 2004) or Argentina (Hebelen et al. 2015). The standardization implies that test plants are cultivated under controlled conditions (temperature, humidity, etc.) before exposure and pots can be placed at the required monitoring sites.

Biomonitoring previously was attributed to field sampling. Use of transplanted plants (in the form of moss bags for example or pots) can reduce quality assurance problems as more uniform plant material can be used and duration of exposure can be fixed.

1. 7. Plant morphologies effect on accumulation

Plant leaf morphology have an effect on the accumulation property of plants (e.g. Li et al. 2012; El-Khatib et al. 2020). The ability of the vegetations to capture and retain airborne particulate matter is directly depends on the interaction between the particulate matter and the plant leaf surfaces. In general, differences in bioaccumulation capacity can be explained by leaf morphological traits such as surface-to-volume ratio (plants with high surface-to-volume ratio accumulate more organic air pollutants than species with compact leaves). Leaves with a larger surface area are more effective in accumulating organic pollutants then those with a smaller area (Desalme et al. 2013).

According to Weerakkody et al. (2018), individual leaf size, shape and morphology show a significant impact on capturing and retaining all particle size fractions. Smaller leaves showed a greater capacity to capture and retain particles probably due their larger parameter per area. Palmately-lobed leaves showed high PM levels compared to elliptical or linear leaves as they

probably create more turbulence in the boundary air layer with their complex shape and "tip-like" areas (Weerakkody et al. 2018).

However, pine needles have found to show higher accumulation properties despite having small leaf area, and being without hairs (Sæbø et al 2012). This may be due to longer narrow needles which may be more easily affected by particulate matter through the thin boundary layer. Pine needles also have their ability to capture larger quantities of PAHs because they can also accumulate PM in winter when the pollution concentration is the highest (Sæbø et al 2012). Roughness, and the presence or absence of leaf hairs may also influence the uptake of PAHs from atmospheric particulate material. It has been reported that leaves exhibiting pubescence (hairiness) had significantly higher total PAH concentrations than hairless leaves (Howsam et al. 2000). Since PAHs are hydrophobic compounds, airborne PAHs are deposited on foliar surfaces mainly by dry deposition (gaseous and particulate-bound forms) (Desalme et al. 2013).

Surface stereo micro-structure and hydrophobicity could have effects on the foliar uptake of organic pollutants. Hairy leaves and leaves with rough ridged surfaces with epicuticular wax were good at capturing and retaining particles in all size fractions compared to leaves with a smooth surface (Weerakkody et al. 2018).

Epicuticular wax content is also an important factor in the accumulation capability of leaves. Margenat et al. (2018) found that relatively high lipid content assists accumulation. PAHs, being lipophilic organic pollutants are expected to accumulate in leaves which contain more extractable lipids such as epicuticular wax + tissue lipid. Davidson and Wu (1990) found poor ability of leaves of *B. cordifolia* to capture and retain particulate matter due to their glossy smooth surface, as particulate matter can easily rebound from smooth surfaces causing only a small percent of captured particles.

The total particulate matter accumulation on leaf within all PM size fraction shows as an increase with the increase of the quantity of leaf wax (Saebo et al. 2012). Studies found reduced PM accumulation on waxy surfaces because of their variable chemical structure and composition and do to their self cleaning ability.

The interspecies differences in PAH concentrations can not be explained by normalizing them to the plant lipid content. PAH in the inner tissues became concentrated with the increase of tissue lipid content while a generally negative correlation between the PAH concentration in cuticles and the epicuticular wax content was found (Qingqing et al. 2017).

2. Main Objectives

Several studies have suggested that the vegetation accumulates atmospheric organic pollutants such as PAHs in leaves (e. g. De Nicola et al., 2011; Bartons et al., 2016) and methods have been developed to use plant foliage as passive samplers to monitor organic pollutants (e. g. Wetzel & Doucette, 2015; Gong et al., 2018). Field bioaccumulation studies might provide useful data (e.g. Pandey et al., 2012; Wang et al., 2017), but these studies lack basic information such as spatial or temporal distribution of contaminants or duration of exposure. During my research the main goal was investigate if a standard protocol, the No. 227 OECD GUIDELINE FOR THE TESTING OF CHEMICALS: Terrestrial Plant Test: Vegetative Vigour Test, can be adopted for bioaccumulation studies and assess the level of air pollution via bioaccumulation studies and to identify possible pollution sources at each sampling site, in parallel with conventional source appointment methods.

The aim of the researches in this dissertation were compare accumulated PAH profiles in different test plants.

Accordingly the main objectives were:

i. to investigate under laboratory conditions the accumulation of PAHs from aerosol, establishing stressor–effect relationships in lettuce (*Lactuca sativa* L., family Asteraceae).
ii. to compare bioaccumulative potential of 8 different vegetables under controlled conditions.
iii. to assess the level of air pollution via bioaccumulation studies in small, medium sized villages in Veszprém County, Hungary in pot experiments using lettuce (*Lactuca sativa* L., family Asteraceae)
iv. to measure PAH levels in *Plantago* samples collected from different sampling sites and in test plants treated experimentally under laboratory conditions and to identify possible pollution sources at each sampling site, in parallel with conventional source appointment methods.
v. to examine and establish the applicability of *C. comosum* for indoor bioaccumulation studies

3. Materials and methods

3. 1. Sample Collection and Sample Preparation

PM samples were collected from two different sources as follows.

I. For testing accumulation of vegetables under laboratory conditions: PM samples were collected with a high-volume KÁLMÁN $PM_{2.5}$ sampler (Figure 2 a) (flow rate 32 $m^3 h^{-1}$) on quartz filter (diameter: 150 mm) (Figure 2 b) from the exhausts of a Euro4 diesel-powered jeep. The sampling was 4 times 10 minutes at idling about 1 meter from the tailpipes.

Figure 2. a.) high-volume KÁLMÁN PM2.5 sampling unit and b.) quartz filter after 10 minutes sampling

II. For testing accumulation of lettuce (Lactuca sativa) under laboratory conditions: PM2.5 aerosol samples were collected on glass fiber filters between 02.12.2016 and 28.12.2016 in Budapest (Hungary) at Gilice square with a high volume Digitel (DHA-80) sampler. The sampling time was 24 hours. This sampling site is located in a suburban area of the Budapest, it is maintained by the Hungarian Meteorological Service. Filters were stored in a freezer at -20 °C until use.

From the filters composite samples were made and the cut filters were extracted by high purity (MilliQ) water (Teke et al. 2020; Kováts et al. 2021a). These pieces were stirred several times, then the beaker was covered for 24 hours (Figure 3 a.). The extract was then filtered on 0.45µm pore size quartz filter (Advantec QR-100 Ø 150 mm) (Figure 3 b.).

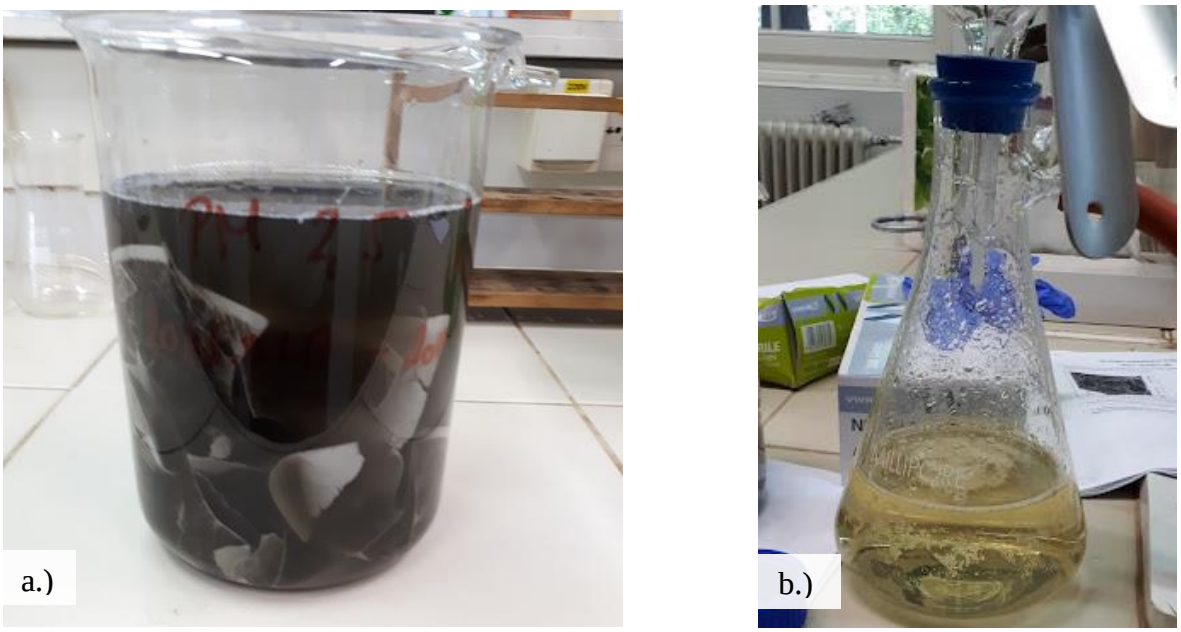

Figure 3. Preparing extract a.) cut filters extract in high purity (MilliQ) water and b.) filtered on 0.45µm pore size filter

3. 2. Cultivation and treatment of test plants

3. 2. 1. Cultivation and treatment of test plants under laboratory conditions

For studying the accumulation of leafy vegetables plant species with relatively high foliar surface have been selected. Finally eight common kitchen garden species were selected which are easy to cultivate also under laboratory conditions. The used species:

- *Lactuca sativa* L. 'Kobak' (lettuce) (Family Asteraceae)
- *Eruca sativa* Mill. (rocket) (Family Brassicaceae)
- *Lepidium sativum* L. (garden cress) (Family Brassicaceae)
- *Apium graveolens* var. secalinum Alef. (leaf celery) (Family Apiaceae)
- *Valerianella locusta* L. (corn salad) (Family Caprifoliaceae)
- *Beta vulgaris* subsp. *vulgaris* convar. *cicla* L. 'Lukullus' (chard) (Family Amaranthaceae)
- *Spinacia oleracea* L. 'Matador' (spinach) (Family Amaranthaceae)
- *Ocimum basilicum* 'Compact' (basil) (Family Lamiaceae)

Cultivation and treatment of test plants based on the No. 227 OECD GUIDELINE FOR THE TESTING OF CHEMICALS: Terrestrial Plant Test: Vegetative Vigour Test recommends several crop species for testing (Annex 2). Seeds of species and/or cultivars used in our studies were purchased from Garafarm Ltd. unless specified otherwise.

The experiment was conducted in a glasshouse; environmental conditions were in concordance with the prescriptions of the Guideline (temperature: 22°C±10°C; humidity: 70% ±25%; photoperiod: minimum 16h light; light intensity: 350±50 μE m^{-2} s^{-1}). Pots were repositioned every other day, therefore potential effect of variations in environmental conditions could be minimised. During exposure, neither fertilisers nor pesticides were applied. The following validity criteria were applied during cultivation and exposure:

The seeds were sown in commercial soil (pH: 6.8 ± 0.5; N (m/m%): min 0.3; P_2O_5 (m/m%): min 0.1; K_2O (m/m%): min 0.3). Pots of 15 cm diameter were used. Prior to exposure, 1 seedling was selected in each pot, others were carefully removed. A control group was set, including 10 pots, similarly to treated groups. Before testing 10 plants were selected and treatment was started when plants reached the 4 true leaf stage,. During the experiments the emergence of the seedlings were min. 70 %. There were no visible phytotoxic effects in the control (e.g. chlorosis, or morphological abnormalities) and at least 90 % mean survival of test plants in the controls.

For each treatment group, 10 replicates (pots) were set. Pots were individually labelled. Control plants were sprayed with tap water simultaneously with the treatments. Layout of the test groups was as follows, indicating repeated treatments Day 0, Day 7, Day 14, Day 21. Treatment implied that test substance was sprayed on the above-ground parts of the plants, using a CONXIN Q1P-CX01-380 portable electric paint spray gun. Application volume was set to 5 mL/pot/treatment. Before spraying, the soil in each pot was carefully covered to avoid contamination and exposure via the roots.

After exposure, the plants were cut above the cotyledon as close as possible to the planting medium and each individual plant was measured using analytical balance (Kern ABJ 120-AM) to four decimal places. As such, biomass was reported as fresh weight. Visual symptoms (if any) were also recorded and rated.

3. 2. 2. Cultivation and treatment of test plants during field experiments

I. Lettuce (Lactuca sativa) pot study

One of the most popular varieties of lettuce “Május királya” (King of May) was used for the test. 10 seeds were sown in commercial soil (pH: 6.8 ± 0.5; N (m/m%): min 0.3; P_2O_5 (m/m%): min 0.1; K_2O (m/m%): min 0.3). Prior to the test, the seedlings were grown in a greenhouse for 40 days. At every sampling site five replicates (5 pots) were used, in one plastic pot with 31 cm diameter five seedlings were planted. During the study period, neither additional fertilizers nor pesticides were used. Exposure took 2 months, between 29.03.2019. and 31.05.2019., after this period the pots were collected and samples were taken to the laboratory where the plants were washed with ultra pure water and stored at -20 °C until analysis.

3. 3. Description of sampling sites and field sampling

I. Lettuce pot study

Seven small/medium-sized Transdanubian villages (Eplény, Hajmáskér, Hárskút, Litér, Nagyvázsony, Pécsely, Tihany) in Veszprém County were selected for the study (Figure 4).

In Eplény, 2 sampling spots were used, in this village a main road (Nr.82) cuts through. On this main road average 8000 vehicles per day were counted, 6500 were light-duty and 1500 were heavy-duty vehicles (https://internet.kozut.hu/kozerdeku-adatok/orszagos-kozuti-adatbank/forgalomszamlalas/). At this sampling sites the gardens which are close to the road might be severely affected. To investigate this assumption one sampling spot was located very close to the road, and the other in the same property, app. 80 meters away from the road.

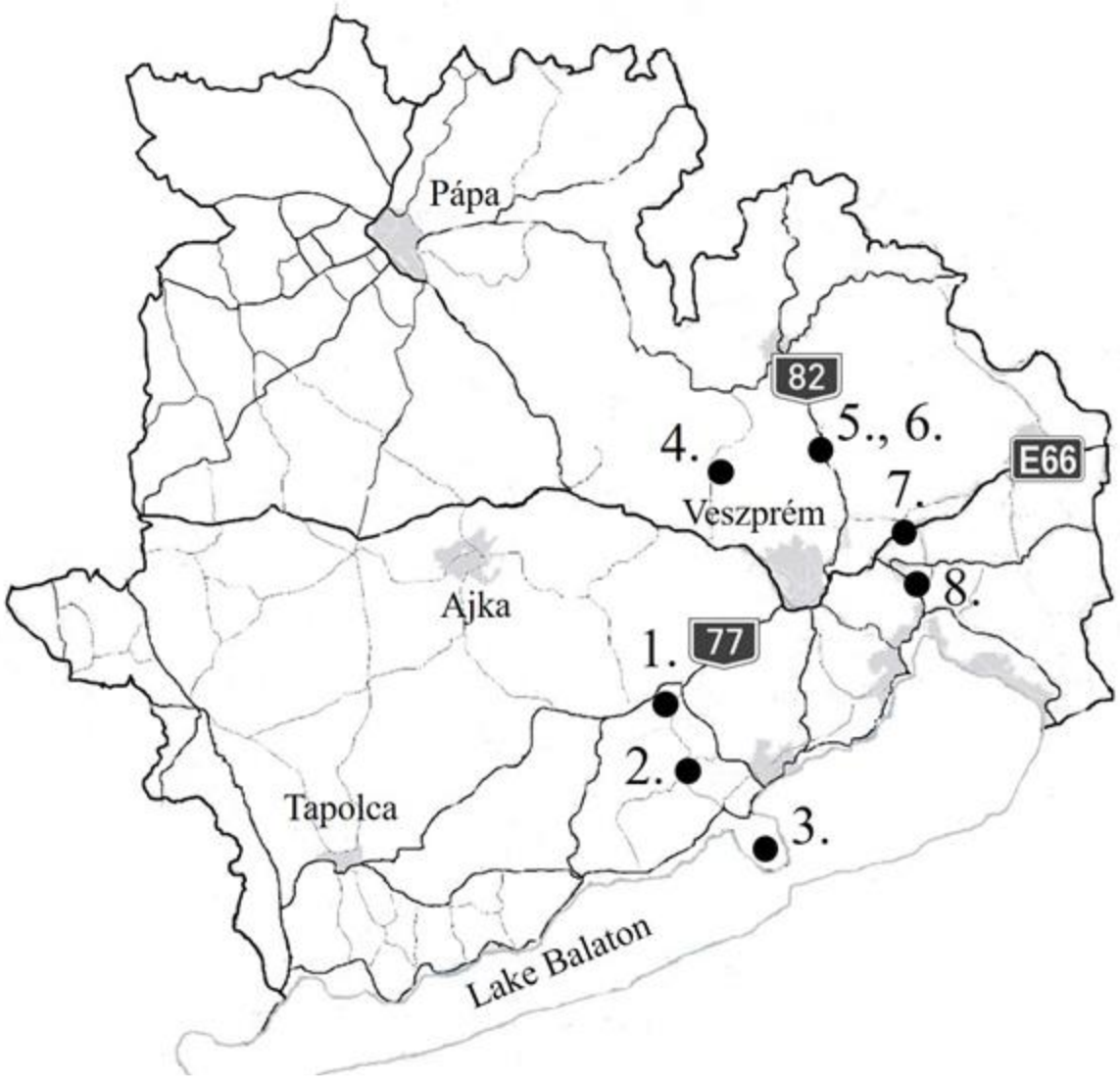

Figure 4. Location of the sampling sites. 1: Nagyvázsony; 2: Pécsely; 3: Tihany; 4: Hárskút: 5: Eplény1; 6: Eplény2; 7: Hajmáskér; 8: Litér.

II. Plantago field collection

Seven sampling sites were selected, this points represent different land use pattern, pollution sources and/or different levels of pollution with special regard to different levels of traffic (Figure 5). The samples were collected on 01.07.2021. Parallel with field sampling laboratory experiment was designed.

Field sampling

Veszprém is the chief town of Veszprém county, it is the biggest town in the county, the population is 58,153 (in 2021). In this city two sampling points were designated, one close to

the central bus station. It has heavy bus traffic from 4:00-23:40. The other point was at a major road cutting through the town, near a petrol fuel station.

Ajka is the third biggest town in Veszprém county, the population is 26.963 (in 2021). In Ajka two sampling points were selected, one was close to the biomass thermal power plant, the other with medium traffic located in the town centre.

Nagyvázsony is a medium-sized village, the sampling spot is situated next to the main road Nr. 77.

In Eplény the sample was collected at the central bus stop by a main road Nr. 82.

Pécsely was selected as background, it is a small village with app. 520 inhabitants. The sampling site was in a relatively unpolluted areas in the Balaton National Park free from human activities and far from any traffic.

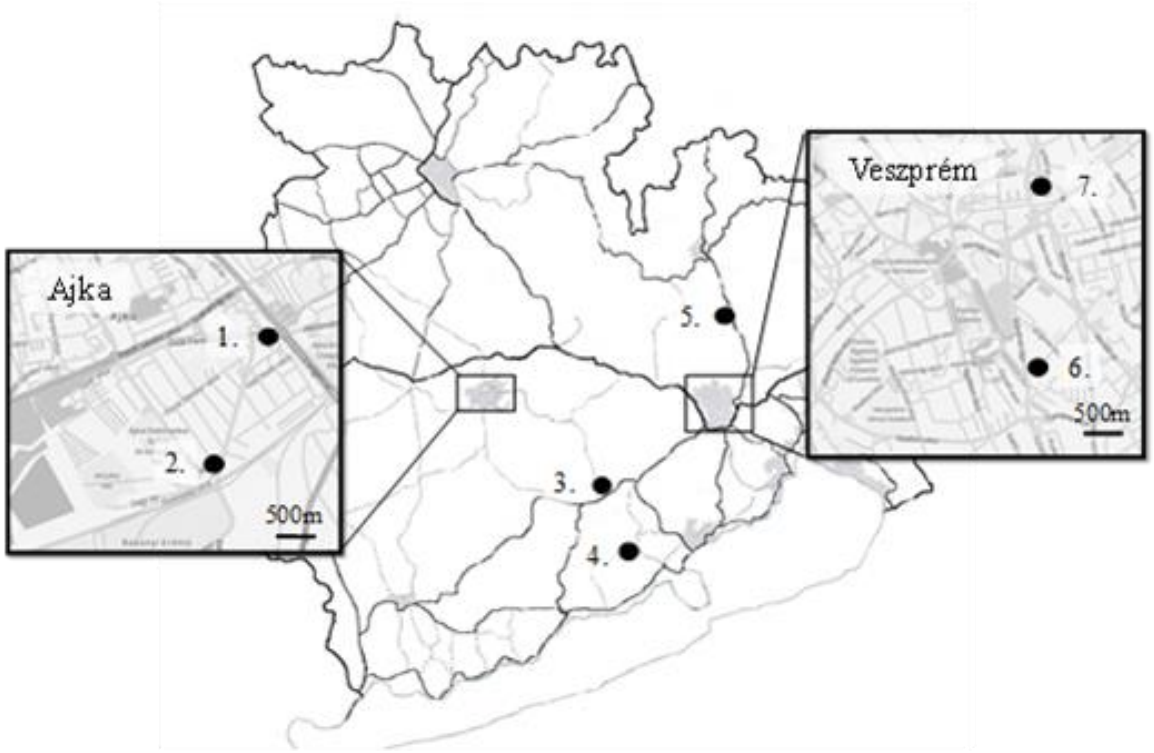

Figure 5. Location of the sampling sites: 1.: Ajka Centre; 2.: Ajka power plant; 3.: Nagyvázsony roadside; 4.: Pécsely National Park; 5.: Eplény bus stop, 6. Veszprém petrol station; 7.: Veszprém bus station

At the selected locations, leaves of three to four fully grown plants of *P. lanceolata* were collected. Leaves were immediately taken to the laboratory, washed with ionic load-free water and frozen (−20◦C) until analysis.

Experimental treatment

P. lanceolata plants were experimentally treated following the No. 227 OECD GUIDELINE FOR THE TESTING OF CHEMICALS: Terrestrial Plant Test: Vegetative Vigour Test.

Organic *P. lanceolata* seeds were purchased from Szentesimag Ltd. 25 seeds were sown in pots of 15 cm diameter in commercial soil (pH: 6.8 ± 0.5; N (m/m%): min 0.3; P_2O_5 (m/m%): min 0.1; K_2O (m/m%): min 0.3). For the test, 3 uniform plantlets were kept in each pot. Cultivation of the test plants and further testing were conducted in a glass-house, environmental conditions were set following the prescriptions of the Guideline (temperature: 22°C±10°C; humidity: 70% ±25%; photoperiod: minimum 16h light; light intensity: 350±50 μE m^{-2} s^{-1}).

For treatment, aqueous extract of PM was used which was collected from a 13 years old, Euro4 diesel-powered jeep. The extract was applied by spraying the sample on the surface of test plants, exposure started when the plants reached the 4- true leaf stage. In contrary to the Guideline which recommends only one spraying at the beginning of the exposure, three treatments were applied: first treatment on Day 0, followed by a second treatment one week later, on Day 7, and then by the third treatment on Day 14. The test was terminated on Day 21. After exposure, leaves were immediately taken to the laboratory, washed with ionic load-free water and frozen (−20°C) until analysis.

A control was also set, where plants received foliar spraying with tap water on the same day as treated plants. Both the control and the treated plant series included 10-10 replicates (10-10 pots).

III. Indoor study

Chlorophytum comosum 'Variegatum' plants with similar size were bought from a local retainer and they were replanted in uncontaminated commercial soil and placed in a greenhouse for four weeks to acclimatize before the study. Four plants were placed in each selected kitchen.

The experiment was two month long, the exposure started 1st of June in 2021 and ended 31th July in 2021. In this period the potential cross-pollution from heating could be avoided because the normal heating season in Hungary is from October until April. The other reason why this period was chosen is that the school holiday starts in June, during summer holidays children mostly lunch at home, which increases cooking frequency.

The leaves were cut after one month (1 month old leaves) and after the second month (2 month old leaves). Leaves were immediately taken to the laboratory, washed with ionic load-free water and kept in the freezer (−20°C) until analysis.

Household selection

Four households were selected with app. the similar size (2 adults + 2 children) (Table 3), which are situated in small villages and not affected by heavy traffic. This criterion was especially important as some indoor biomonitoring studies reported that in case of traffic-impacted sites such as schools, infiltration of outdoor air pollutants could be experienced.

Table 3. Key data for the surveyed households

	HH1	HH2	HH3	HH4
Number of inhabitants	4	4	4	4
Cooking frequency per day	1, very seldom 2 (less than 5%)	usually 2	usually 2	1, very seldom 2 (less than 5%)
Energy source	electric stove/oven	gas stove electric oven	electric stove/oven	electric stove/oven
Material used	lard app. 40% vegetable (sunflower) oil app.55% olive oil app.5%	lard app. 95% butter app. 5%	lard 1% vegetable (sunflower) oil app. 89%, coconut app. 10%	vegetable (sunflower) oil app.100%
Cooking method	Cooking 35% Deep-frying 30% Pan-frying 15% Oven 20%	Cooking 40% Deep-frying 0% Pan-frying 40% Oven 20%	Cooking 30% Deep frying 55% Pan frying 5% Oven 10%	Cooking 50% Deep-frying 15% Pan-frying 5% Owen 30%
Ventilation	poor	good	good	good

3. 4. Analysis of Polycyclic aromatic hydrocarbons (PAHs)

Analytical determinations of the PAHs in the aerosol filters water solutions and, plants, were performed in the testing laboratory at the Laboratory of the ELGOSCAR-2000 Environmental Technology and Water Management Ltd. accredited by the National Accreditation Authority, registration number NAH-1-1278/2015. The concentrations of PAHs in the samples were measured by a gas chromatograph-mass spectrometer (GC-MS) instrument.

The analysis was carries out for 16 priority PAHs (EPA–PAHs) which the US Environmental Protection Agency (EPA) enlists posing the highest environmental risk. (reviewed by Keith 2015). The following EPA-PAHs compounds were measured naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo[k]fluoranthene, benzo(a)pyrene, benzo(e)pyrene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene, benzo(g.h.i)perylene.

3. 4. 1. Sample preparation of PAHs in Plant material

Ten grams of plant sample was grinded with 10 g anhydrous sodium sulphate in a ceramic mortar. The extraction was repeated 3 times with 20 ml n-hexane in ultrasonic extractor for 20 min (Figure 6 a). Prior to extraction, 10 ml acetone was added and the samples were spiked with 100 µl of 0.01 µg ml^{-1} deuterated PAH surrogate mixture (naphtalene-d8, acenaphthene-d10, phenantrene-d10, chryzene-d12, benzo(a)pyrene-d12, and perylene-d12). The extract was dehydrated with anhydrous sodium-sulphate and concentrated with dry nitrogen stream at 40°C to a 1 ml.

In order to reduce the amount of the matrix compounds from the plant extract which could interfere with the analysis an additional solid-phase silica gel and alumina oxide sample clean-up was performed. 1ml of the concentrated sample was passed through a 30 cm long, 0.5 cm width glass column containing 0.2 g silylated (deactivated) glass wool, and 3 g of silica gel on the bottom of the column, and 3 g of aluminum oxide on the top, and was washed with 20ml of n-hexane, after this washing step ~~and~~ it was eluted with 20-ml methylene chloride (Figure 6 b., c., d.). The column was washed with 20 ml n-hexane. The solid phase cleanup provides a much cleaner sample where most of the plant matrix like lipids and pigments like chlorophyll A and B are separated form the analytes.

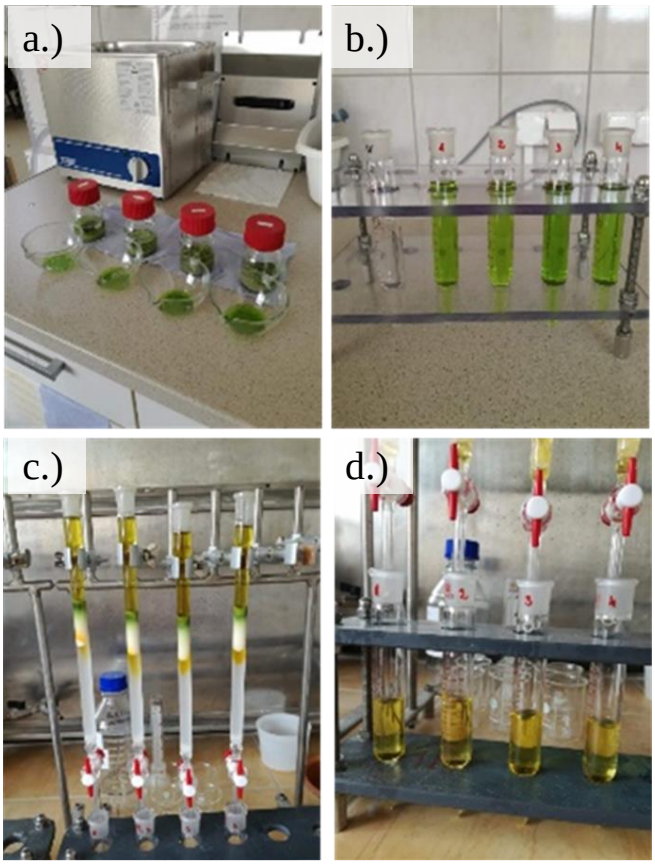

Figure 6. Plant sample laboratory analysis procedures. a.) pretreated plant samples and sample extracts. b.) plant samples n-hexane extracts prior to nitrogen stream concentration. c.) solid phase clean-up of plant extracts. d.) cleaned sample eluted in methylene chloride prior to concentration.

The clean sample in methylene chloride was concentrated with dry nitrogen stream at 40 °C to 1 ml. Before the measurement the remaining samples were transferred into amber colored analytical vials and 100 µl of 0.01 µg ml^{-1}, internal standard mixture (2-floro-biphenyl, and p-terphenyl-d14) was added (final concentration was 100 µg kg^{-1} plant dry wt).

The plant samples were analyzed by Agilent 6890GC 5973E MSD GC-MS based on MSZ (Hungarian Standard) EN 15527:2009. Under the conditions specified in the standard, limit of detection of 0.1μg/kg for each individual PAH can be achieved.

3. 4. 2. Sample preparation of PAHs from aerosol filters water solution

For measuring PAH content of aerosol filters water solution samples, the MSZ (Hungarian Standard) 1484-6: 2003: Environmental protection, Testing of waters. Part 6: Determination of polycyclic aromatic hydrocarbons (PAH) content by gas chromatographic-mass spectrometry was followed. Under the conditions specified in the standard, typical limit of detection is 0.001 μg L^{-1} water sample. Gas chromatographic-mass spectrometric method Hungarian standard was followed. 250ml of water sample was extracted in a 500 ml separation funnel with 10 ml n-hexane; the extraction was repeated two times. Prior to extraction, the samples were spiked with 50 μl PAH surrogate mixture (deuterated, naphthalene D8, acenaphthene D10, phenanthrene D10, chrysene D12, benzo(a)pyrene D12, perylene D12) (Table 4) the final concentration was 1 μg L^{-1} water.

The extract was dehydrated with anhydrous sodium-sulphate and concentrated with dry nitrogen stream at 40 °C to a 1 ml. An additional solid-phase silica gel and alumina oxide sample clean-up was performed. The column was washed with 20 ml hexane and the extract was eluated with 20 ml dichloromethane. This extract was concentrated with dry nitrogen stream at 40 °C to a 1 ml, and the dissolvent was change to hexane. 100 μl of 0.01 μg ml^{-1}. Internal standard mixture (2-floro-biphenyl, and p-terphenyl- d14) was added (final concentration was 4 μg L^{-1} water). Before the analysis the remaining samples were transferred into amber colored analytical vials.

3. 4. 3. GC-MS analysis of PAHs

The GC-MS analysis was performed with an HP-6890 gas chromatograph; it was coupled to an HP-5973 (Agilent Technologies, Palo-Alto, USA) quadrupole mass spectrometer (low-resolution single MS). Injector and transfer-line temperatures were 320 °C and 250 °C, respectively, and ion source and quadropool temperatures were 280 °C and 150 °C, respectively. A spitless glass liner, 4 mm i.d., loosely filled with silanized glass wool at the bottom was used (Restek) in the split/splitless GC injector (320 °C, purge splitless 1.5 min). The GC column was 30 m × 0.25 mm i.d., film thickness 0.25 μm, ZB-Semivolatiles (Phenomenex). The GC oven temperature was maintained at 40 °C for 3 min after injection

then programmed at 40 °C min^{-1}, 40 to 80 °C for 0.5 min, and then at 15 °C min^{-1} to 240 °C which was maintained for 8 min and then at 15 °C min^{-1} to 310 °C and which was maintained for 8 min. Helium (N55) was used as carrier gas at 1.2 mL min^{-1} in constant flow. The acquisition mode was SIM (single ion monitoring). Electron ionization was used with energy of 70 eV. The figure below shows the analyte compounds structure and mastered SIM ions.

3. 4. 4. Calibration of PAHs

A five-level internal calibration was used. The first level was the limit of quantitation and the last level was the 80% of the linear range. During the calibration each calibration level was prepared form certified reference materials with dilution. During the calibration each compound and surrogate standards relative response factors was determined for every calibration level. The compounds quantitation ion responses were tabulated agents each target analyte and internal standard concentrations. The compounds Response factors (RF) was calculated with the following equation:

$$\boldsymbol{Rfi = (Ai * Cs)/(As * Ci)}$$

Where: Rf_i = Response factor of analyte i

Ai = Peak response of compound i

Cs = Concentration of internal standard (mg kg^{-1}, or mg L^{-1})

As = Peak response Internal Standard

Ci = Concentration of analyte or surrogate (mg kg^{-1}, or mg L^{-1})

3. 4. 5. Qualitative analysis of PAHs

The PAH compounds was identified based on their previously determined GC-MS retention times and their previously gathered mass spectrometric selective ion monitoring (SIM) target ion and qualifier ion ratios. In order to qualify a specific PAH, the analyte retention time and the SIM ion ratios has to match with the certified reference materials parameters (Table 4).

Table 4. Measured compound qualification data: names, retention times, SIM target and qualifier ions and ion ratios.

Analyte name	CAS - number	Retention time t_R (min)	Target ion (m z^{-1})	Qualifier ion (m z^{-1})	ratio (%)
Naphthalene-D8 Surrogate standard	1146-65-2	8.59	136	134	10.1
Naphthalene	91-20-3	8.62	128	127	10.1
2-methylnaphthalene	91-57-6	9.76	142	141	83.1
1-methylnaphthalene	90-12-0	9.89	142	141	89.8
Acenaphthylene	208-96-8	11.16	152	153	13.1
Acenaphthene-D10 Surrogate standard	15067-26-2	11.38	164	162	100.2
Acenaphthene	83-32-9	11.43	154	152	89.5
Fluorene	86-73-7	12.30	166	165	97.2
Phenanthrene-D10 Surrogate standard	1517-22-2	13.77	188	184	14.8
Phenanthrene	85 -01-08	13.80	178	176	15.5
Anthracene	120-12-7	13.91	178	176	14.8
Fluoranthene	206-44-0	15.82	202	200	19.6
Pyrene	129-00-0	16.25	202	200	20.3
Benzo(a)anthracene	56-55-3	20.01	228	229	20.0
Chrysene-D12 Surrogate standard	1719-03-5	20.10	240	241	18.8
Chrysene	218-01-9	20.14	228	229	19.3
Benzo(b)fluoranthene	205-99-2	25.68	252	250	23.4
Benzo(k)fluoranthene	207-08-9	25.78	252	250	22.5
Benzo(e)pyrene	192-97-2	26.51	252	250	28.5
Benzo(a)pyrene-D12 Surrogate standard	63466-71-7	26.59	264	260	6.7
Benzo(a)pyrene	50-32-8	26.66	252	250	23.6
Perylene-D12 Surrogate standard	1250-96-3	26.83	264	260	2.5
Indeno(1,2,3-cd)pyrene	193-39-5	29.29	276	278	3.0
Dibenzo(a,h)anthracene	53-70-3	29.41	278	276	31.3
Benzo(g,h,i)perylene	191-24-2	29.82	276	278	3.8
2-fluorobiphenyl Internal standard	321-06-8	10.33	172	171	25.6
p-terphenyl-D14 Internal standard	1718-51-0	16.65	244	245	19.8

3. 4. 6. Quantitative analysis of PAH samples

During the quantitative analysis an internal standard method was used. When using an Internal standard calibration, a known amount of non-target compounds is added to the sample post-extraction but prior to analysis. After peak identification the analytes and the integral standards peak area is measured by integration. Two internal standards were used 2-floro-biphenyl, and p-terphenyl- d14. The goal for the addition of internal standards is corrects for response changes due to instrument drift and matrix effects.

The compounds concentration was calculated with the following equation:

$$C_i=(A_i*C_s*V_s) / (Rf_i*A_s*V_x)$$

Where: Ci = Concentration of analyte or surrogate (μg kg^{-1}, or μg L^{-1})

A_i = Peak response of compound i

C_s = Concentration of internal standard (μg kg^{-1}, or μg L^{-1})

Vs = standard amount

Rf_i = Response factor of analyte i

A_s = Peak response Internal Standard

V_x = sample amount

In order to evaluate the effectiveness of the sample preparation and to correct the matrix effect and the analyte losses during sample preparation surrogate standards was added before sample preparation. Surrogate standards where analyzed a same as the rest of the analytes. The surrogate standard recovery was determined. In order the surrogate standards to mimic the chemical properties of PAH analytes stabile isotope labelled deuterated PAH surrogate mixture containing Naphthalene-d8, Acenaphthene-d10, Phenanthrene-d10, Chryzene-d12 Benzo(a)pyrene-d12, and Perylene-d12 was used.

3. 5. Quality control of PAH analysis

Because analysis was carried out in a certified accredited laboratory the quality control measures where in harmony with the laboratory accreditation to ISO/IEC 17025:2018, and with the laboratory's internal quality management system guidelines. During the methods development the analytic performance has been evaluated. The following Method performance parameters has been checked: Selectivity, Precision (Repeatability), Accuracy, Linearity, Limit of detection (LOD). Limit of quantifications (LOQ).

The PAHs analysis Repeatability and Accuracy was measured by the repeated analysis of spiked samples. The PAH plant samples RSD ranged from 5.47% (Chrysene) to 17.51% (Naphthalene). The average analyte recovery for Plant spiked PAH samples ranged from 88.3% (Benzo(k)fluoranthene) to 106,4 (Anthracene). In the aerosol filter water extract spiked samples RSD ranged from 2.11% (Benzo(g,h,i)perylene) to 10.74% (Naphthalene). The average analyte recovery for spiked aerosol filter water extract samples ranged from 93.7% (Phenanthrene) to 111.2% (1-methylnaphthalene). 1. Annex contains the Repeatability and Accuracy values of PAH analytes in spiked plant samples. 2. Annex contains the Repeatability and Accuracy values of PAH analytes in aerosol filter water extract spiked samples.

The LOD values 0.01μg kg^{-1} to 0.015μg kg^{-1} for accumulated PAH in plant samples and 0.00015 μg l^{-1} to 0.0003μg L^{-1} in aerosol filter water extract samples. The LOQ values in plant samples range from 0,031 μg kg^{-1} to 0.05μg kg^{-1} and 0.00029 μg L^{-1} to 0.001μg L^{-1} in aerosol filter water extract samples. 3. Annex contains LOD and LOQ values of PAH analytes in spiked plant samples. 4. Annex contains LOD and LOQ values of PAH analytes in aerosol filter water extract spiked samples.

Blank sample was analyzed with each sample batch. The sample preparation of the Blank samples was following the same procedures of the actual samples described above. According to EPA standards the blank sample was accepted when the target analyte concentration was less than half of the quantifications. For the reduction blank concentrations and to prevent cross contamination, the glassware was thoroughly cleaned before use, washing with non-ionic detergent and rinsing with ultrapure water and n-hexane, and dried in oven at 150 °C.

Quality control (QC) samples was prepared with each sample batch and analyzed before and after the samples. The QC samples was made by adding a known amount of analytes to sample matrix.

In each batch the recovery of the PAHs with in the QC samples has been measured. The QC samples where accepted when the analyte recoveries fall between 80% to 120% of the calculated amount. If the values exceeded the acceptance range the whole batch would have been re analyzed.

3. 6. Source appointment

Ratios of specific PAH compounds, such as fluoranthene to the sum of fluoranthene and pyrene (FLT/(FLT + PYR)), benzo[a]anthracene to the sum of benzo[a]anthracene and chrysene (BaA/(BaA + CHR)), and indeno[1,2,3-cd]pyrene to the sum of indeno[1,2,3-cd]pyrene and benzo[g.h.i]perylene (IPY/(IPY + BPE)), were calculated to evaluate the possible sources of PAHs in the samples.

3. 7. Statistical analysis

The correlation between BCF and molecular weight was calculated with Spearman's rank correlation and the correlation between tested vegetables and individual PAHs were calculated with Pearson correlation. In order to examine compositional differences among samples,

principal component analysis (PCA) has been performed which generally reduces the set of variables into two major principle components. PCA has been extensively used to evaluate PAH accumulation pattern in different plant matrices (e.g. Kodnik et al. 2015; Capozzi et al. 2017). Statistical analyses were performed using the RStudio (RStudio Desktop 1.4.1106) programme ggfortify package (https://CRAN.R-project.org/package=ggfortify) and R 4.0.0programme (http://cran.r-project.org/src/base/R-4/R-4.0.0.tar.gz) Rcmdr package. Accumulated amounts of PAHs in plants were compared using Spearman correlation; coefficients were determined to estimate the dependence of sampling site on the levels of PAHs found in Plantago samples. To identify the relationship between the PAH content of samples and sampling sites, PCA and factor analysis were performed with RStudio.Statistical significance was defined as $p \leq 0.05$.

4. Results and discussion

4. 1. Foliar uptake and accumulation of airborne polyaromatic hydrocarbons using lettuce test plants.

4. 1. 1. Accumulated PAHs amount in lettuce plants

Totally 19 PAHs were found after the analysis in lettuce sample, of them 16 were priority PAHs enlisted by US EPA. From the EPA PAHs the only exception was dibenzo(a,h)anthracene which was not detected in our plant samples. Naphthalene was determinated in highest concentration 72 μg kg^{-1} and 2-methyl-naphthalene was in lower amount 22.85 μg kg^{-1} in treated lettuce samples. The second highest accumulated amount of phenanthrene was found 15.55 μg kg^{-1}. The lowest amount of acenaphthylene was determined 2.6 μg kg^{-1}. Table 5 shows the composition of the PM2.5 aerosol extract and the concentration of the accumulated compounds in lettuce.

Table 5. Concentration of PAHs in the aerosol extract and in the lettuce leaves. Bioconcentration Factors (BCFs) are also indicated. Priority PAHs are given in italic bold. (LOQ: Limits of quantification)

PAH	PM2.5 sample [μg L^{-1}]	Treated lettuce [μg kg^{-1}]	BCF	Molecular weight [g mol^{-1}]
Naphthalene	0.39	72	184.61	128.17
2-methyl-naphthalene	0.19	22.85	120.26	142.20
1-methyl-naphthalene	0.15	11.55	77.00	142.20
Acenaphthylene	0.02	2.6	130.00	152.19
Acenaphthene	<LOQ	1.05	<LOQ	154.21
Fluorene	0.04	3.55	88.75	166.22
Phenanthrene	0.39	15.55	39.87	178.23
Anthracene	0.03	6.15	205.00	178.23
Fluoranthene	0.59	12.15	20.59	202.25
Pyrene	0.59	11.4	19.32	202.25
Benzanthracene	0.15	7.45	49.67	228.29
Chrysene	0.27	7.6	28.14	228.30
Benzo(b)fluoranthene	0.22	11.35	51.59	252.31
Benzo(k)fluoranthene	0.07	3.45	46.28	252.31
Benzo(e)pyrene	0.1	3.7	37.00	252.32
Benzo(a)pyrene	0.09	4.55	50.56	252.32
Indeno1,2,3CD-Pyrene	0.12	4	33.33	276.33
Benzo(g,h,i)perylene	0.08	2.8	35.00	276.30
Dibenzo[a,h]anthracene	<LOQ	<LOQ	<LOQ	278.35
Total PAH	3.53	205.5	58.22	-

4. 1. 2. Calculated bioconcentration factor in lettuce

Bioconcentration Factor (BCF) was calculated according to the following ~~the~~ equation (Kacálková and Tlustoš 2011):

$$BCF = \frac{\text{PAH concentration in treated lettuce [μg L} - 1]}{\text{PAH concentration in the PM2.5 sample [μg kg} - 1\ ]}$$

Remarkably high BCFs were experienced for naphthalene (184.61) and for anthracene (205). In the aqueous extract of PM2.5 the concentration of naphthalene was already relatively high (0.396 μg L^{-1}), significantly higher than in a winter urban PM10 sample (Kováts et al. 2017). As a general rule, as particulate matter size decreases, relatively more potentially toxic compounds are bound; e.g. Valavanidis et al., (2006) reported that the amount of PAH on fine

particles were higher than on coarse ones. The lowest BCF (19.32 and 20.59, respectively) was given by pyrene and fluoranthene. No PAHs were detected in the control plants.

The BCF for uptake and accumulation of PAHs in most studies are calculated from soil (e.g. Zohair et al. 2005; Zhang et al. 2015; Inam et al. 2016). Mo et al. (2009) for example report that highest BCF of total PAHs (5.5) was found in *Brassica* sp. collected in the Pearl River Delta (South China). Khan and Cao (2012) calculated RCFs (root/soil concentration factor) and SCFs (shoot/soil concentration factor) for different vegetables grown in metropolitan areas of Beijing (China) and found that with the increase of ring numbers the bioaccumulation factors decreased. Regardless, several studies have shown that atmospheric deposition is the major pathway for the accumulation of PAHs in vegetation (Jia et al. 2019). Li et al. (2008) for example was not able to find correlation between total PAHs in vegetable samples with soil samples. Some data are available, however, on the relationship between atmospheric concentration of PAHs and their accumulation in higher plants. Sharma and Tripathi (2009), found that BCFs of total PAHs in the leaves of *Calotropis gigantea* (an evergreen shrub) were in the range of 1.00 - 11.72, while BCF for total PAHs was 58.22 in our experiment. However, it is difficult to compare different species due to differences in important attributes such as leaf morphology or life cycle (Franzaring and van der Eerden 2000).

Studies showing PAH accumulation specifically in lettuce reports a quite wide range of concentration values. In a pot experiment of Gelman (2014), practically there was no accumulation detected in rooftop gardens in Helsinki. In another study, Jia et al. (2018) found that the total concentrations of 16 PAHs in samples collected from industrial areas of Shanghai ranged between 132.0-319.2 μg kg^{-1}. Concentration of total PAHs in exposed lettuce plants in our study was 205.5 μg kg^{-1}, which falls into this range, indicating highly polluted conditions.

In our experiment strong correlation was found between BCF and molecular weight (Spearman's rank correlation: p= 0.009, S= 1315.5, rho= -0.6121725). The lower molecular weight (LMW) PAH compounds were more dominant after the treatment, which is in consistency with other studies (e.g. Lei et al. 2011; Wang et al. 2017; Jia et al. 2018). In my experiment in treated lettuce samples the concentration of naphthalene was the highest (72 μg kg^{-1}), it was also reported to be one of the dominant PAH in bioaccumulation studies (Busso et al. 2018). As a general rule, the highly lipophilic PAH molecules (heavy PAHs) showed lower accumulative potential than the less lipophilic ones (light PAHs) (Paraíba et al. 2010).

4. 2. Foliar uptake and accumulation of polycyclic aromatic hydrocarbons from diesel emissions.

4. 2. 1. Accumulated amount of PAHs in test plants

In the plants and extract altogether 15 PAHs were found, all of them belonging to the group of the US EPA enlisted 16 priority PAHs (Table 6). The detected PAHs included all carcinogenic PAHs. In the extract the most dominant PAHs were phenanthrene (0.264 μg L^{-1}), fluoranthene (0.22 μg L^{-1}) and pyrene (0.121 μg L^{-1}). The highest accumulated concentration was detected in leaf celery (total PAH: 31.9 μg kg^{-1}) despite the fact that only 5 different PAHs could be determined. Phenanthrene was the most dominant 19.30 μg kg^{-1} and pyrene was in the lowest concentration. The lowest concentration of accumulated PAHs was measured in Basil (total PAH: 9.2 μg kg^{-1}) and also this was also the plant with the fewest types of PAHs were bonded in leaves.

Table 6. PAHs concentration in the diesel extract and in the plant samples. US EPA priority PAHs are given in bold. Carcinogenic PAHs are given in Bold. LOQ: Limit of quantification

PAH	Diesel extract [µg L^{-1}]	Lettuce [µg kg^{-1}]	Rocket [µg kg^{-1}]	Chard [µg kg^{-1}]	Leaf celery [µg kg^{-1}]	Spinach [µg kg^{-1}]	Garden cress [µg kg^{-1}]	Corn salad [µg kg^{-1}]	Basil [µg kg^{-1}]
Acenaphthylene	0.022	0.80	1.20	1.00	3.00	1.30	<LOQ	0.90	<LOQ
Fluorene	0.034	1.90	1.80	0.80	5.80	1.40	<LOQ	0.70	1.20
Phenanthrene	0.264	7.00	6.80	0.55	19.30	8.30	5.70	4.10	6.80
Anthracene	0.018	0.30	0.40	0.40	<LOQ	0.40	<LOQ	0.30	<LOQ
Fluoranthene	0.220	7.20	0.50	2.30	2.30	3.20	1.60	1.90	1.20
Pyrene	0.121	5.40	2.10	1.50	1.50	2.00	1.10	1.50	<LOQ
Benzo(a)anthracene	***0.019***	***1.50***	***1.10***	***1.00***	***<LOQ***	***0.80***	***1.10***	***0.70***	***<LOQ***
Chrysene	***0.032***	***2.50***	***1.50***	***0.60***	***<LOQ***	***0.70***	***<LOQ***	***0.90***	***<LOQ***
Benzo(b)fluor-anthene	***0.036***	***1.40***	***1.40***	***0.90***	***<LOQ***	***0.70***	***<LOQ***	***1.30***	***<LOQ***
Benzo(k)-fluoranthene	***0.013***	***0.50***	***0.60***	***0.20***	***<LOQ***	***0.30***	***<LOQ***	***0.30***	***<LOQ***
Benzo(e)pyrene	0.018	0.50	0.30	0.40	<LOQ	0.30	<LOQ	*0.30*	<LOQ
Benzo(a)pyrene	***0.007***	***0.40***	***0.50***	***0.30***	***<LOQ***	***<LOQ***	***<LOQ***	***0.40***	***<LOQ***
Indeno1,2,3CD-Pyrene	***0.008***	***0.30***	***0.40***	***0.20***	***<LOQ***	***<LOQ***	***<LOQ***	***0.30***	***<LOQ***
Dibenzo[a,h]-anthracene	***0.001***	***<LOQ***	***<LOQ***	***<LOQ***	***<LOQ***	***0.60***	***<LOQ***	***<LOQ***	***<LOQ***
Benzo(g,h,i)-perylene	***0.005***	***0.30***	***<LOQ***	***0.30***	***<LOQ***	***<LOQ***	***<LOQ***	***<LOQ***	***<LOQ***
TOTAL PAH	0.818	30.0	18.60	10.45	31.9	20.0	9.5	13.6	9.2

4. 2. 2. Bioaccumulation potential in vegetables

Figure 7 shows the concentration of different molecular weight PAHs groups in the tested vegetables. The highest bioaccumulation potential was shown by leaf celery, the measured concentration of PAHs was 31.9 µg kg^{-1}. In a pot study De Temmerman et al. (2012) found high accumulate potential for atmospheric heavy metals, but no data were found about the celery leaves PAH accumulation capacity. They used two consecutive years and the

exposure periods were 2 months. The highest bioaccumulative potential was shown for Pb reaching as high as 6757 mg kg^{-1} fresh weight.

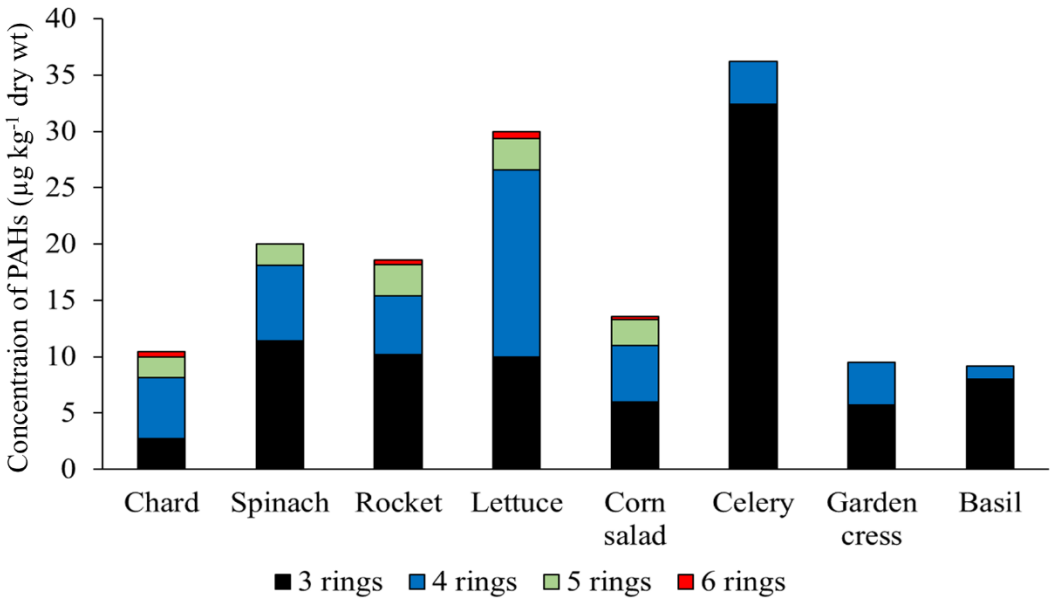

Figure 7. Concentration of different molecular weight PAHs groups in the tested vegetables

In our study lettuce (*L. sativa)* accumulated approximately 30.0 µg kg^{-1} ΣPAHs. Lettuce has shown generally higher bioaccumulation capacity in previous studies than other vegetables (Li et al. 2015; Mombo et al. 2016), this species has high foliar surface and thin cuticula which make it a good accumulator (Schreck et al., 2012).

The accumulation of spinach and rocket were similar, the PAH concentration in spinach was 20 µg kg^{-1} and in rocket 18.6 µg kg^{-1}. Pandey et al. (2012) found that spinach had good bioaccumulation capacity. In pot experiment the spinach leaves accumulated to the highest extent from atmosphere and to lower extent from irrigation water through the roots (Pandey et al. 2012). Field studies also showed the risk of PAH contamination in leafy vegetables. Abou-Arab et al. (2014) investigated the concentration of PAHs in vegetable leaves collected from different regions in Cairo (Egypt). They detected high amounts of carcinogenic PAHs except pyrene, the highest concentration of total PAHs was measured in spinach (8.977 µg kg^{-1}) (Abou-Arab et al., 2014).

Jia et al. (2018) collected samples near industrial areas of Shanghai and they found in vegetables that the total concentrations of 16 PAHs ranged from 65.7 to 458.0 µg kg^{-1}. In the leafy vegetables the highest accumulation was in spinach (*Spinacia oleracea* var.) (223.3-458.0 µg

kg^{-1}), followed by Chinese cabbage (*Brassica rapa* var.), (206.0-348.1 μg kg^{-1}), Shanghai green cabbage (*Brassica chinensis* var.), (206.4-284.7 μg kg^{-1}), and finally lettuce (*Lactuca sativa* L.) (132.0-319.2 μg kg^{-1}).

There is no information about bioaccumulation potential of *E. sativa* despite it is a very popular herb in urban gardening. In several studies other Brassicaceae species have been used in bioaccumulation experiments such as Collard greens (*Brassica oleracea* var. *acephala*) (Amato-Lourenco et al. 2016), Chinese cabbage (*Brassica chinensis* L.) (Yang et al. 2022), pakchoi (*Brassica campestris*) (Wang et al. 2018a) and Shanghai green cabbage (*Brassica rapa subsp. chinensis*) in the above-mentioned study of Jia et al. (2018). In general, cabbage is found to be a good accumulator of PAHs (Xiong et al. 2017; Zhang et al. 2018).The total PAHs concentration in corn salad was 13.6 μg kg^{-1}. Ultimately chard, garden cress and basil showed the lowest accumulation rate, in decreasing order: 10.45 μg kg^{-1}, 9.50 μg kg^{-1} and 9.20 μg kg^{-1} total PAHs.

Cultivation of garden cress and basil is even recommended in the so-called balcony gardens, which can be in the proximity of roads. Up to now, there have been very insufficient amount of information of the PAH accumulative capacity of these species. Taking into consideration *O. basilicum*, most studies address the accumulation of heavy metals, which appears to be lower in comparison to other vegetables (Patrick-Iwuanyanwu and Chioma 2017). Laboratory scale studies also showed that heavy metal uptake is significant via the roots (Adamczyk-Szabela et al., 2017).

During our study different plant families were represented. However, in one genus significant differences can be found, e.g. in the investigation of Mo et al. (2009) the amount of 16 PAHs were measured in vegetable species which were collected close to Pearl River Delta, South China on cultivated area. Three species belonged to the genus *Brassica* and ∑PAHs concentration varied as following (mean values are given): flowering Chinese cabbage (*Brassica parachinensis*) 438 μg kg^{-1}, Paitsai (*Brassica chinensis*) 950 μg kg^{-1} and Mustard (*Brassica juncea*) 1790 μg kg^{-1}.

Uptake of organic pollutants can even vary amongst varieties (Zohair et al. 2006). Li et al (2017) investigated three plant species *Hypericum, Photinia,* and *Mahonia* in a very small sampling area, which had similar air pollution level, they found that the ΣPAHs uptake by the plant leaves showed significant differences among the species.

Basically, differences in bioaccumulation capacity can be explained by the differences in the leaf morphological traits such as surface-to-volume ratio (Franzaring and van der Eerden, 2000). Epicuticular wax content also plays an important factor (Li et al. 2017). Leaf roughness, and the presence or absence of leaf hairy structures can also influence the uptake of PAHs from atmospheric particulate material (Amato-Lourenco et al. 2017).

The differences in the bioaccumulation potential however, can only be partly explained by morphological traits. Of the three species showing the lowest bioaccumulation rate, namely chard, garden cress and basil, chard and garden cress have rather thick epidermis while basil has not (Bozokalfa et al., 2016; Raklevičienė et al., 2007).

4. 2. 3. Bioaccumulation pattern of PAH compounds

Not only the bioaccumulation potentials of tested vegetables were different also the accumulation pattern (Figure 8). Good correlation was found between the PAH content of the shoots and that of the diesel extract in most investigated vegetables (Table 7), the best correlation was found in lettuce ($t = 12.536$, $df = 13$, p-value <0.05)

Table 7. Pearson correlation between tested vegetables and individual PAHs

Plant species	correlation coeff. (Pearson)	p- values
Apium graveolens* var. *secalinum	0.9299	<0.01
Beta vulgaris* subsp. *vulgaris* convar. *cicla	0.5952	<0.05
Eruca sativa	0.7229	<0.01
Lactuca sativa	0.9610	<0.01
Lepidium sativum	0.8709	<0.01
Ocimum basilicum	0.7931	<0.01
Spinacia oleracea	0.9121	<0.01
Valerianella locusta	0.9185	<0.01

Leaf celery accumulated only 3- and 4-ring PAHs, despite this, it had the highest accumulation rate (%). Accumulation only of these PAHs were observed in cress and basil, which showed the lowest accumulation potential, but the pattern was quite different: garden cress accumulated a relatively large amount of 4-ring PAHs and basil, prevalence of 3-ring PAHs was found (Figure 8).

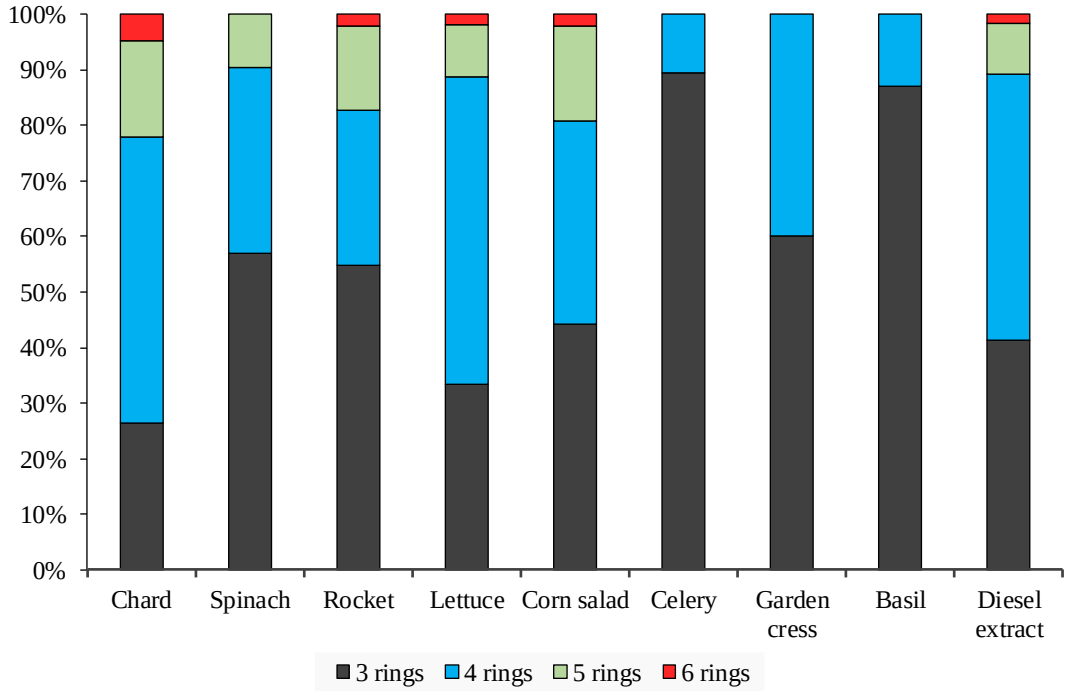

Figure 8. Percentage contribution of different molecular weight PAHs groups in the tested vegetables

Spinach accumulated only 3-, 4-, and 5-ring PAHs, while the other vegetables tested accumulated all PAHs present in the extract. The relative contribution of 6-ring PAHs was the highest in Swiss chard. The lettuce, rocket, corn salad and chard accumulated all type of PAHs which were present in the extract. Relative contribution of 6-ring PAHs was the highest in chard.

In our experiment 3 and 4 ring low molecular weight (LMW) PAH compounds were predominant in all vegetables (Figure 8), which was consistent with other studies (e.g. Wang et al. 2017; Jia et al. 2018). Rodriguez et al. (2015) measured the amount of accumulated PAHs in a pot study of *Lolium perenne* exposed with vehicular emissions, they detected no high molecular weight (HMW). According to the Chinese study of Jiang et al. (2015) LMW PAH compounds were dominant in vegetable oils. Phenanthrene had the highest concentration in all tested vegetables and reaching as much as 19.3 µg kg^{-1} in leaf celery, 8.3 µg kg^{-1} in spinach, 7.0 µg kg^{-1} in lettuce, 6.8 µg kg^{-1} in rocket and basil, 5.7 µg kg^{-1} in garden cress and 4.1 µg kg^{-1} in corn salad. In our previous pot study in which we measured the accumulation of lettuce plants at different locations with varying level of traffic, in all samples phenanthrene had the second highest concentrations (11.6 µg kg^{-1}) and also fluoranthene and pyrene was found in high concentrations (7.2 and 5.4 µg kg^{-1}, respectively).

4. 3. Accumulation of airborne polyaromatic hydrocarbons in lettuce samples in rural gardens

4. 3. 1. Accumulation in test plants

From the analyzed 19 PAHs only acenaphthylene, acenaphthene, anthracene, and dibenzo[a,h]anthracene were not detected in the plant samples. Table 8 summarizes the analytically measured concentrations and detect rates of all PAHs in lettuce leaves. The highest amount of PAHS were detected in samples from Hajmáskér (186 μg kg^{-1}) and less than the half of this total PAH concentration was measured in Hárskút (80 μg kg^{-1}). In these samples naphthalene was the most dominant PAH (155 and 32.7 μg kg^{-1}). The lowest accumulated total PAH concentration was determinated in lettuce samples from Litér (9.1 μg kg^{-1}). In this sample only six different types of PAH were find, these are naphthalene, 2-methyl-naphthalene, 1-methyl-naphthalene, fluorine, phenanthrene, fluoranthene. In contrast, in the sample with the highest accumulated amount ten types of PAH were identified naphthalene, 2-methyl-naphthalene, 1-methyl-naphthalene, fluorine, phenanthrene, fluoranthene, pyrene, benzanthracen, benzo(b) fluoranthene, benzo(e)pyrene. The highest number of different types of PAH was found in samples from Hárskút, totally 15.

Table 8. Concentration of PAHs in lettuce samples. LOQ: Limit of quantification

PAH compounds	PAHs concentration (µg kg^{-1} dry weight) in lettuce leaves							
	Pécsely	Nagyvá-zsony	Eplény 2	Eplény 1	Tihany	Litér	Hárskút	Hajmáskér
Naphthalene	15.4	9.76	3.94	41.9	12.7	3.1	32.7	155
2-methyl-naphthalene	1.6	5.6	1.5	2	2.5	1.5	5.2	6
1-methyl-naphthalene	1.6	2.2	<LOQ	<0.01	1.3	<0.01	2.8	2.5
Acenaphthylene	<LOQ	<LOQ	<LOQ	<0.01	<0.01	<0.01	<0.01	<0.01
Acenaphthene	<LOQ	<LOQ	<LOQ	<0.01	<0.01	<0.01	<0.01	<0.01
Fluorene	<LOQ	1.1	<LOQ	1.3	1.1	1.1	1.6	1.9
Phenanthrene	11.6	3.4	4.9	5.4	5.5	2.2	11.1	9.9
Anthracene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
Fluoranthene	2.1	2.9	11.2	3.4	1.7	1.2	4.3	3.1
Pyrene	1.8	1.7	7.3	1.9	1.1	<LOQ	3.4	2.5
Benzanthracene	1.2	<0.01	1.6	<LOQ	1.3	<LOQ	3.9	1.6
Chrysene	1.5	1.2	4	1.2	1	<LOQ	2.8	<LOQ
Benzo(b) fluoranthene	1.7	<LOQ	5.1	1.1	1.6	<LOQ	4.2	1.1
Benzo(k) fluoranthene	<LOQ	<LOQ	1.2	<LOQ	<0.01	<LOQ	1.6	<LOQ
Benzo(e)pyrene	1.1	<LOQ	1.1	<LOQ	1	<LOQ	1.9	1.4
Benzo(a)pyrene	1.2	<LOQ	2.3	<LOQ	1	<LOQ	2	<LOQ
Dibenzo[a.h] anthracene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<0.01	<LOQ
Indeno1.2.3CD-Pyrene	<LOQ	<LOQ	1.6	<LOQ	<LOQ	<LOQ	1.4	<LOQ
Benzo(g.h.i) perylene	<LOQ	<LOQ	1.9	<LOQ	<LOQ	<LOQ	1.1	<LOQ
Total PAHs	40.8	27.9	47.6	58.2	31.8	9.10	80.0	186

Figure 9 shows the total amount of different molecular weight PAHs on the sampling spots. In all the test sites, the low molecular weight (LMW) PAH compounds were dominant in exposed lettuce plants (Figure 9), which is consistent with other published studies (Lei et al. 2011; Wang et al. 2017; Jia et al. 2018). The accumulated concentration of naphthalene was in the range

between 3.1 µg kg^{-1} (Litér) and 155 µg kg^{-1} (Hajmáskér), with fairly high concentrations in Hárskút (32.7 µg kg^{-1}) and Eplény1 (41.9 µg kg^{-1}). Naphthalene is in general one of the dominant PAHs in bioaccumulation studies (Waqas et al. 2014; Busso et al. 2018).

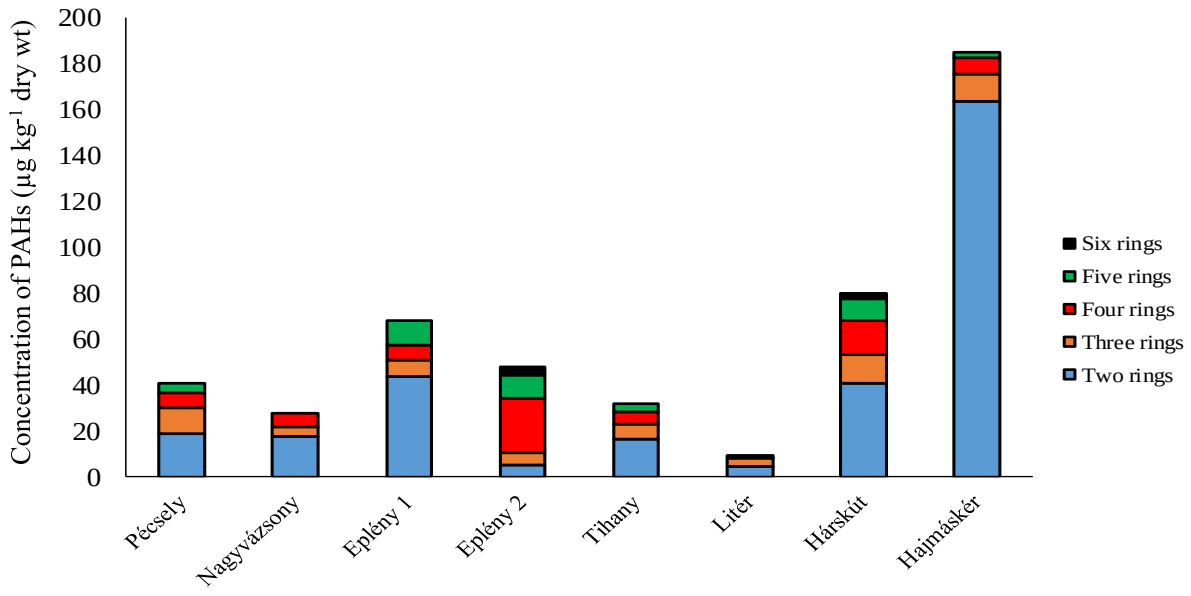

Figure 9. Total amount of different molecular weight PAHs in plants in µg kg^{-1} dry weight

In plant samples the most abundant PAH compound was phenanthrene, the concentration was between 2.2 to 11.6 µg kg^{-1}. In the study of Li et al. (2008) the PAH content of 30 agricultural soil and 16 vegetables were determined and phenanthrene was found one of the most dominant PAH in vegetables.

The detection rate of anthracene was LOD, though this PAH showed high levels in all vegetable samples (including spinach and cabbage) procured from local wholesale markets in Punjab (Pakistan) (Ashraf et al. 2013).

The highest concentration of the 4-ring PAHs, pyrene and the carcinogenic chrysene were in Eplény2 (7.3 and 4.0 µg kg^{-1}, respectively), followed by Hárskút (3.4 and 2.8 µg kg^{-1}).

Relative abundance of the five-ring PAHs was rather high in 4 samples (Pécsely, Eplény2, Tihany, Hárskút) showing peaks in Eplény2 and in Hárskút. The identified five ring-PAHs, benzo(k)fluoranthene, benzo(b)fluoranthene and benzo(a)pyrene are considered as typical tracers for fossil fuel combustion and are associated with vehicular emissions (Ravindra et al.

2008). The contribution of these PAHs ranged from 88 % and 80 % of this group in Eplény2 and in Hárskút.

Of the six-ring PAHs, only two, namely indeno1.2.3.CD-pyrene and benzo(g,h,i)perylene were detected only in Eplény2 and Hárskút samples. Their total concentration was 3.5 μg kg^{-1} in Eplény2 and 2.5 μg kg^{-1} in Hárskút. According to Eiguren-Fernandez et al. (2004), benzo(g,h,i)perylene is considered a marker of gasoline exhaust emissions. Analysis of individual land-use patterns might give some explanation: Hárskút is a village which is situated in a mountainous area and is a Natura2000 site. However, there are several farms in the neighbourhood of the sampling site and the high pollution can be caused by the inputs of fossil fuel from farm vehicles which generally pose high environmental load (Ene et al. 2012). Eplény2 sampling site is in the proximity of the railway, thus diesel-powered engines might be responsible for the pollution.

The total PAH concentration was ~~the~~ in the range of 9.1 μg kg^{-1} dry.wt (Nagyvázsony) and 185 μg kg^{-1} dry.wt (Hajmáskér) (Table 9). In related literature, the amount of accumulated PAHs shows very diverse values. Several reports have shown low accumulated amount of PAHs in leafy vegetables, Abou-Arab et al. (2014) e.g. measured 8.9 μg kg^{-1} total PAH in spinach (*Spinacia oleracea* var.) which were collected from the urban region of Cairo (Egypt). Also low accumulation was reported in spinach in urban gardens of Sao Paulo (Brasil) by Amato-Lourenco et al. (2017), in their experiment only one garden was monitored for PAH accumulation, where the PAH concentration was 7.4 μg kg^{-1}. Data reported by Jánská et al. (2006) make regional comparison possible. In their study, the accumulation of total PAHs ranged between 12.34 and 78.09 μg kg^{-1} in cabbage samples collected from Southern Moravia (Czech Republic). Even lower accumulation was found in a Romanian study (Soceanu et al. 2014), where maximum total PAHs accumulated concentration ranged to 8.216 μg kg^{-1} in cabbage and 8.131 μg kg^{-1} in spinach collected from rural areas.

However, much higher values were observed in industrial areas of Shanghai, here the total accumulated amount of 16 PAHs in spinach ranged from 223.3-458.0 μg kg^{-1}~~.~~ (Jia et al. 2018).

Nevertheless, taking into consideration that the different vegetables will have different accumulation potential (Franzaring and van der Eerden 2000), comparison of our results with other leafy vegetables have only indicative value. Examining the other studies where lettuce was used as passive monitor, PAH accumulation in lettuce moved in a very wide range. In a

pot experiment by Gelman (2014), practically no accumulation was found in experimental rooftop gardens in Helsinki. On the contrary, in the previously mentioned study of Jia et al. (2018), the total concentrations of 16 PAHs in lettuce collected from Shanghai industrial areas showed accumulation between 132.0-319.2 μg kg^{-1}. The highest value in our study (Hajmáskér, 185 μg kg^{-1} dry.wt) is in fact in this range, indicating significant contamination.

4. 3. 2. PAH concentrations in soil samples

In parallel to vegetable samples, concentration of 19 PAHs, including the 16 EPA PAHs, was measured in all soil samples (Table 10). As a general rule, PAHs have high tendency to accumulate in soils because their strong hydrophobicity and resistance to degradation, it was found that app. 90% of total PAHs retain in surface soils (Wild and Jones 1995). In our study the upper 5 cm layer of the soil was analyzed.

In soil samples from the analyzed PAHs only the amount of acenaphthylene was under the detection limit (Table 9).

In soil samples the highest concentration of accumulated PAHs was determined in samples from Hárskút (595 μg kg^{-1} dry weight), in this samples Fluoranthene was the dominant PAH (107 μg kg^{-1} dry weight). Less than a half of the total amount from Hárskút was measured in sample from Eplény1 (211 μg kg^{-1} dry weight), where the we found the Naphthalene as the dominant PAH (154 μg kg^{-1} dry weight). The lowest total PAH content was identified in plants from Litér (31.6 μg kg^{-1} dry weight), where similar to the sample collected in Hárskút Fluoranthene was the dominant PAH (6.9 μg kg^{-1} dry weight).

Table 9 Concentration of PAHs in soil samples. LOQ: Limit of quantification

PAHs compounds	PAHs concentration (µg kg^{-1} dry weight) in soil samples							
	Pécsely	Nagyvá-zsony	Eplény 2	Eplény 1	Tihany	Litér	Hárskút	Hajmáskér
Naphthalene	13.3	33.1	5.1	154	<0.01	1.1	17.3	15.7
2-methyl-naphthalene	1.5	2.6	1.5	5	1.9	1.2	3.1	1.5
1-methyl-naphthalene	<LOQ	1.2	<LOQ	3.1	1	<0.01	1.9	<LOQ
Acenaphthylene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
Acenaphthene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	4.8	<LOQ
Fluorene	<LOQ	<LOQ	<LOQ	1.6	1	<0.01	4	<LOQ
Phenanthrene	5	6	5.4	13.1	4	2.3	56.5	5.8
Anthracene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	21.8	<LOQ
Fluoranthene	7.4	8.6	10.8	4.7	8.6	6.9	107	8.4
Pyrene	5.5	6	7.2	3.3	5.8	5	86.2	6.1
Benzanthracene	1.8	1.6	2.4	4.5	1.4	1.5	68.2	2.1
Chrysene	1.5	1.6	4	4.4	3.3	1.5	43.3	3.4
Benzo(b) fluoranthene	4.3	4.8	5.9	6	4.2	4.3	57.9	4.7
Benzo(k) fluoranthene	1.3	1.5	1.6	2.1	1.3	1.4	24.3	1.1
Benzo(e)pyrene	1.1	1.1	1.6	2.4	<0.01	1.1	38.7	1.2
Benzo(a)pyrene	2	2	3.3	3.2	1.8	1.5	24.4	2.8
Dibenzo[a.h] anthracene	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	5.3	<LOQ
Indeno1.2.3CD-Pyrene	1.3	1.4	2.3	2.2	1.2	1.1	17.7	2
Benzo(g.h.i) perylene	1.2	1.9	2.2	1.7	1.4	2.7	12.7	2,00
Total PAHs	36.9	73.4	53.3	211	36.9	31.6	595	56.8

Figure 10 shows the total amount of different molecular weight PAHs in the sampling spots.

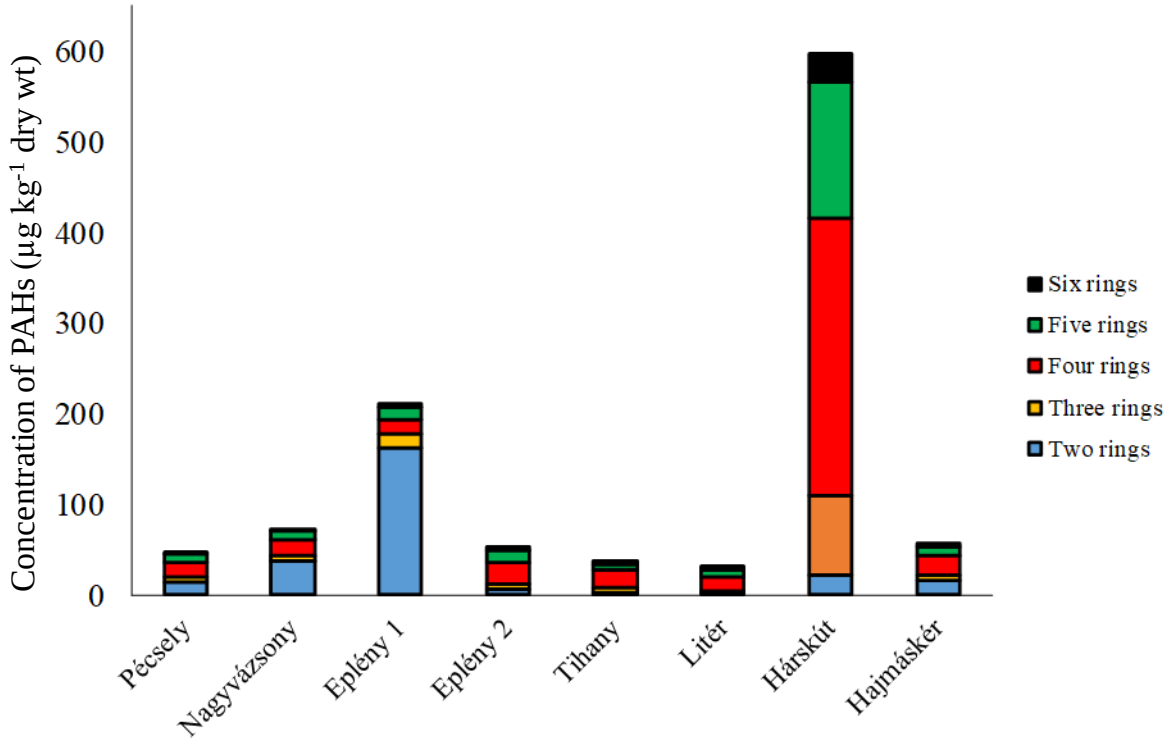

Figure 10. Total amount of different molecular weight PAHs in plants in µg kg^{-1} soil

The total concentration of PAHs was in the range of 31.6 µg kg^{-1} (Litér) and 595.1 µg kg^{-1} (Hárskút). The highest accumulation tendency in the majority of samples was shown by four ring PAHs. The PAH concentrations of these four ring PAHs were in the range between 14.9 µg kg^{-1} (Litér) and 304.7 µg kg^{-1} (Hárskút). Five ring accumulated PAHs, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene and benzo(a)pyrene occurred in all of the samples while the detection rate of dibenzo[a.h]anthracene was much lower, it occurred only in Hárskút, with the concentration of 5.3 µg kg^{-1}. A study by Maliszewska-Kordybach et al. (2009) analysed the spatial distribution of individual PAHs in agricultural soils in over 200 localities in Poland and found that the higher molecular hydrocarbons (groups of 4+5+6 rings PAHs) represented 72.6% of the total PAH content. Also, some Chinese studies reported dominance of HMW PAHs in contaminated soils (Wang et al. 2016; Wang et al. 2017). A study by Bozlaker et al. (2008) which analyzed dry deposition and soil samples indicated that soil acts as the sink of HMW PAHs throughout the year. Another study by Zavgorodnyaya et al. (2019) also showed that wet deposition resulted in the accumulation of HMW PAHs.

The comparison between the individual villages and Hárskút showed outstandingly high concentrations. Concentration of 3 ring PAHs amounted to 87.1 µg kg^{-1}, 4 ring PAHs to 304.7

μg kg^{-1}, 5 ring PAHs to 150.6 μg kg^{-1} and 6 ring PAHs to 30.4 μg kg^{-1}. The amount of individual PAHs was roughly one magnitude higher than in the other samples (Table 10)

A work of Maliszewska-Kordybach (1996) suggested a contamination classification system for rhizosphere soils, defining the following categories: weakly contaminated (> 200 μg kg^{-1}), contaminated (600–1000 μg kg^{-1}), and heavy contamination (> 1000 μg kg^{-1}). Of our samples, Eplény1 and Hárskút fall into the contaminated category, the others are classified as weakly contaminated. This classification system, however, cannot be fully used in this study as soil PAH concentrations refer only to a two-month exposure time. However, the 50/2001. (IV. 3.) Decree of the (Hungarian) Government defines a 1mg kg^{-1} limit value for the PAH content of wastewater sludges intended for agricultural use.

4. 3. 3. Source appointment

In order to identify the possible source of different PAHs, a long established PAH isomer ratios were used which have also been applied to allocate sources of these compounds in vegetables (e.g. Li et al. 2008) and also in soils (e.g. Yang et al. 2017b). A ratio higher than 0.5 for fluoranthene to fluoranthene plus pyrene (Flt/Flt+Pyr) indicates grass, wood or coal combustion as the potential source (Yunker et al 2002). This value was exceeded in all of the plant samples, which shows the contribution of household heating and biomass combustion, most probably also, open burning agricultural waste in the gardens.

Flt/(Flt+Pyr) ratio could be calculated for all vegetable samples, the other ratios provided much less information. Benz[a]anthracene to benzo[a]anthracene plus chrysene (BaA/BaA+Cry) ratio if it is over 0.35 has been defined to indicate combustion of vegetation and fossil fuel (Yunker et al. 2002) (Figure 11). In our study, this ratio could be calculated for only four lettuce samples: Pécsely, Tihany, Eplény2 and Hárskút. In these cases, BaA/(BaA+Cry) ratio was in the range of 0.37 (Eplény2) and 0.5 (Tihany), showing the contribution of pyrogenic sources.

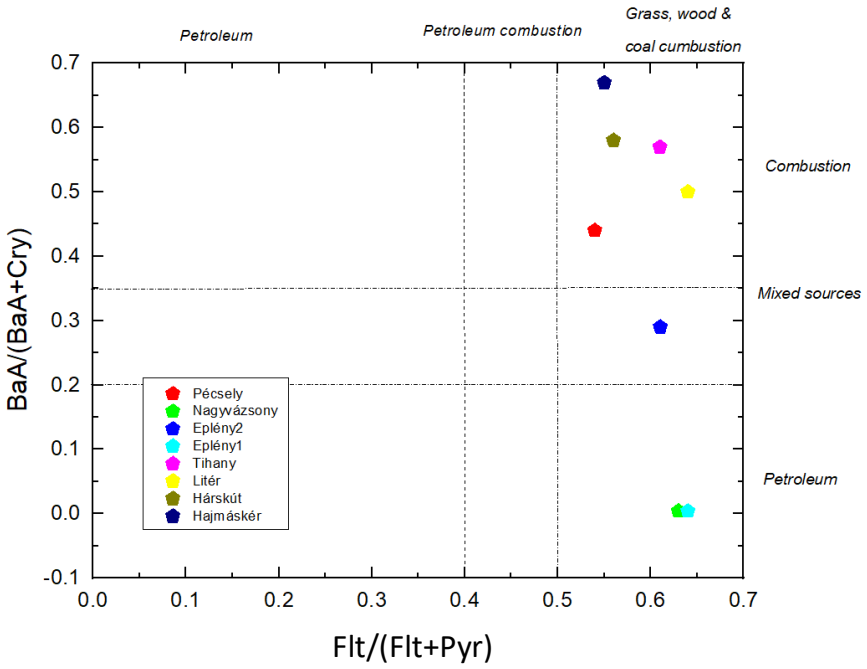

Figure 11. Cross-plot of BaA/(BaA+Cry) ratio against Flt/(Flt+Pyr) for plant samples

Indeno[1,2,3-cd]pyrene to indeno[1,2,3-cd]pyrene plus benzo[g.h.i]perylene (Ind/Ind+BghiP) ratios between 0.20 and 0.50 most likely indicate liquid fossil fuel combustion while ratios lower than >0.50 imply grass, wood and coal burning (Figure 12) (Yunker et al., 2002). Because 6-ring PAHs occurred only in lettuce samples exposed in Eplény2 and Hárskút site, ratios could be calculated only for these samples. In Eplény2, this ratio was 0.46, indicating fossil fuel combustion as the potential source, while in Hárskút it amounted to 0.56, indicating biomass combustion. Considering individual, characteristic PAHs, dibenzo[a,h]anthracene (DbaA) concentration was below detection rate. This compound is typically associated with coal combustion (Pant et al. 2017).

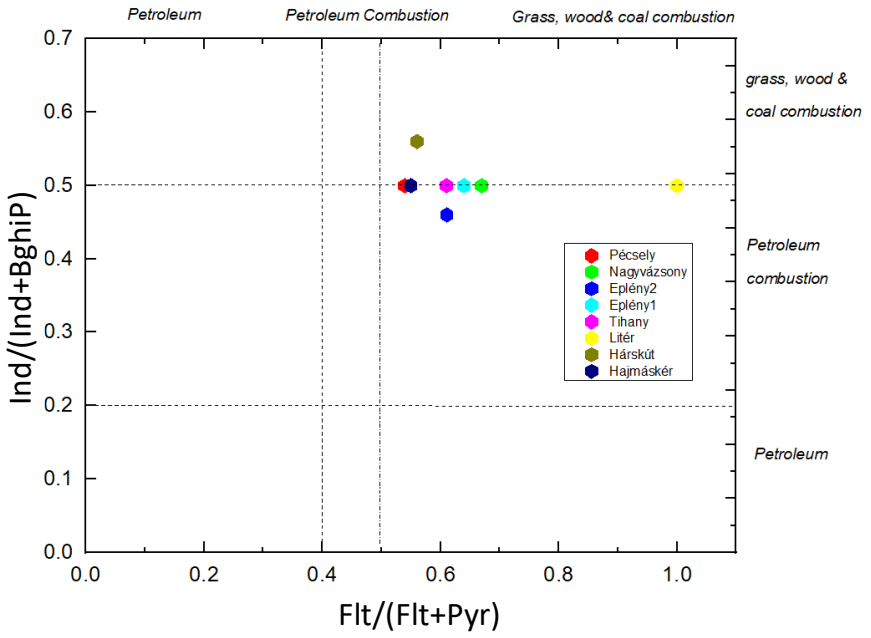

Figure 12. Cross-plot of Ind/(Ind+BghiP) ratio against Flt/(Flt+Pyr) for plant samples

When analyzing the source origin of different PAHs in the soil samples similarities can be found with the lettuce samples. The Flt/(Flt+Pyr) ratio exceeded 0.50, indicating the input from biomass burning, regardless of the size or location of the village. Ind/Ind+BghiP ratios could be calculated for all of the soil samples: values were between 0.20 and 0.50 in case of Nagyvázsony, Tihany and Litér (which indicates liquid fossil fuel usage), at the transition point of 0.5 in Hajmáskér and >0.50 in Eplény1, Eplény2, Hárskút and Pécsely (which indicates biomass burning, most possibly grass and wood) (Yunker et al., 2002).

BaA/(BaA+Cry) ratio was <0.35 in case of only 1 village, Tihany (0.30) which can indicate either petroleum or combustion. In the other sampling sites it was in the range of 0.38-0.61 which implies combustion of vegetation and fossil fuel (Yunker et al., 2002).

For soil samples, BaA/(BaA+Cry) ratio against Flt/(Flt+Pyr) ratio as well as Ind/(Ind+BghiP) ratio against Flt/(Flt+Pyr) ratio were plotted (Figure 13, 14). Plotting BaA/(BaA+Cry) ratio against Flt/(Flt+Pyr) ratio shows high input from combustion in general for the majority of the villages. The cross plot of Ind/(Ind+BghiP) ratio against Flt/(Flt+Pyr) ratio, however, distinguishes two groups of villages: in case of Eplény1 and 2, Pécsely, Hajmáskér and Hárskút

the main input is biomass combustion while in case of Litér, Tihany and Nagyvázsony, petroleum combustion provides the main source.

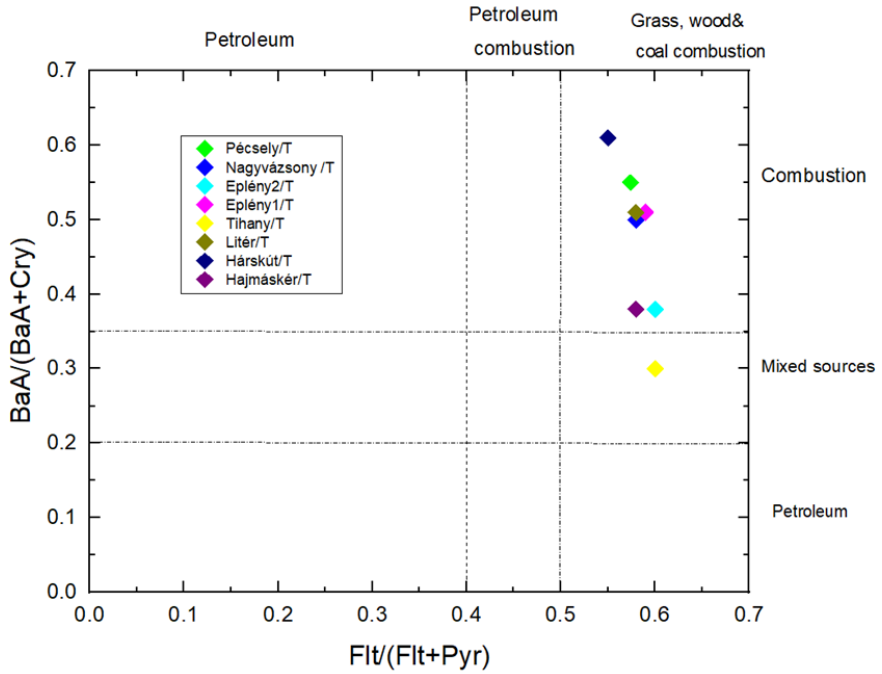

Figure 13. Cross-plot of BaA/(BaA+Cry) ratio against Flt/(Flt+Pyr) for soil samples

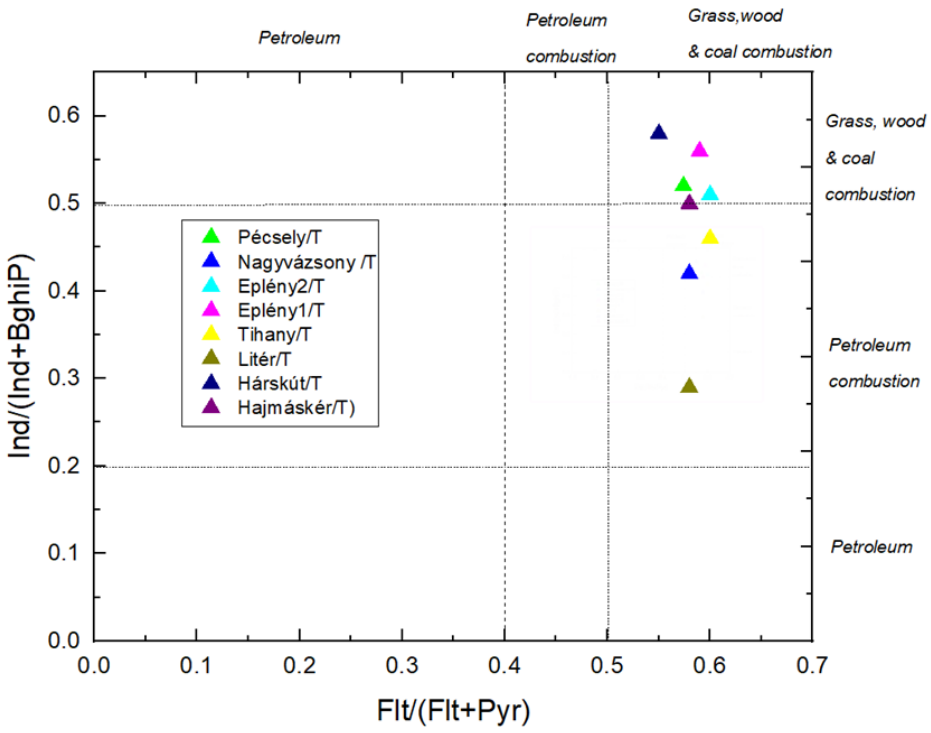

Figure 14. Cross-plot of Ind/(Ind+BghiP) ratio against Flt/(Flt+Pyr) for soil samples

When assessing the two samples collected in Eplény, it is interesting to note that they are having very different composition, though the two sampling spots are situated app. 80 ms from each other. Eplény1 sampling site was especially chosen to represent roadside conditions: approximate load is 8000 vehicles per day, of which 6500 are light-duty and 1500 are heavy-duty vehicles (https://internet.kozut.hu/kozerdeku-adatok/orszagos-kozuti-adatbank/forgalomszamlalas/). In an early study of Larsson and Sahlberg (1981) accumulation of PAHs in lettuce was assessed. Samples were grown at 12 and 50 ms distance from a Swedish highway. Significantly higher accumulation was found at the vicinity of the highway, with the concentration of B(g.h.i.)P 0.5 μg kg^{-1} vs. 10.8 μg kg^{-1}. Dan-Badjo et al. (2007) placed ryegrass (*Lolium perenne*) pots in the vicinity of a highway and found that the high molecular weight PAHs (5 and 6 rings) represented almost 23% of the total PAH concentrations. In a following study, however, spatial distribution of accumulation was depicted (Dan-Badjo et al., 2008). It was recorded that concentration of accumulated PAHs was the highest between 0 and 10 m on both sides of the road than it started to decrease rapidly. However, source appointment did not show extra burden from liquid fossil fuel combustion in Eplény1, though the pots were placed app. 5 metres from the road. In this soil sample, concentration of total PAHs was relatively high (211.3 μg kg^{-1}), but prevalence of two-ring PAHs was experienced, concentration of naphthalene was 154 μg kg^{-1}. Naphthalene was also a dominant PAH in the vegetable sample, with the concentration of 41.9 μg kg^{-1}.

4. 4. PAHs accumulation pattern with *Plantago lanceolata* L. as a passive biomonitor

4. 4. 1. Accumulated PAH amount in treated Plantago lanceolata

In our research Phe, Flt and 4-ring Pyr were found dominant in diesel extract (Table 11) these results are similar to those reported by Fabiańska et al., (2016), Lin et al., (2019) and Corrêa et al (2021). In most of the studies, the ratio of five- or more ring species is very low (Jin et al. 2014; Yilmaz and Davis 2016).

The total PAH amount in treated *P. lanceolata* leaves was 92.2 μg kg^{-1}. B(b)f represented 17% of total individual PAHs, followed by B(a)p (15%), Phe and B(e)p (9%), Ind (8%), B(k)f and B(g,h,i)p (7%), Pyr and Cry (4.5%), Flt (4.4%) and Nap, B(a)a and D(a,h)a (3%). Overall, 5-ring PAHs were dominant, amounting to 50% of total PAHs.

Bioconcentration Factors (BFC) were calculated to determine the accumulation pattern of individual PAHs (Table 10), with BCF the coefficient between the tested organism and the external medium (Paraíba et al., 2010). The following equation was used to calculate the BCF: BCF = PAH concentration in the *P. lanceolata* leaves/PAH concentration in the sample (Kacálková and Tlustoš 2011).

We recognized that carcinogenic PAHs had higher BCF than other congener PAHs, BCF of D(a,h)a was 2660, B(a)p 1971 and IP 886. Lowest BCF was found in case of Flt (18.5) and Ant (23.9), respectively.

Table 10. Concentration of PAHs in the aerosol extract and in the experimentally treated plant samples. Bioconcentration factors (BCFs) and molecular weights are also shown. Car PAHs are given in Bold

PAH	Diesel extract	Plantago treated	BCF	Molecular weight
	[µg L^{-1}]	[µg kg^{-1}]		[g mol^{-1}]
Acenaphthylene	0.022	0.53	24.1	152.19
Fluorene	0.034	1.27	37.4	166.22
Phenanthrene	0.264	8.64	32.7	178.23
Anthracene	0.018	0.43	23.9	178.23
Fluoranthene	0.22	4.08	18.5	202.25
Pyrene	0.121	4.19	34.6	202.25
Benzo(a)anthracene	**0.019**	**2.9**	**152.6**	**228.29**
Chrysene	**0.032**	**4.13**	**129.1**	**228.3**
Benzo(b)fluoranthene	**0.036**	**15.7**	**436.1**	**252.31**
Benzo(k)fluoranthene	**0.013**	**6.38**	**490.8**	**252.31**
Benzo(e)pyrene	0.018	8.24	457.8	252.32
Benzo(a)pyrene	**0.007**	**13.8**	**1971.4**	**252.32**
Indeno1,2,3CD-Pyrene	**0.008**	**7.09**	**886.3**	**276.33**
Dibenzo[a,h]anthracene	**0.001**	**2.66**	**2660.0**	**278.35**
Benzo(g,h,i)perylene	**0.005**	**6.36**	**1272.0**	**276.3**
TOTAL PAH	0.818	92.2	112.7	-

4. 4. 2. Accumulated PAH concentrations in Plantago lanceolata samples collected at different sampling locations

In collected *Plantago* samples the concentration of 19 accumulated individual PAHs in leaves were measured (see Table 11). According to field samples, the lowest total PAH concentration was found at Veszprém petrol station (34.7 µg kg^{-1}), higher amount of accumulated PAHs were detected in sample collected in Ajka close to the power plant (38.6 µg kg^{-1}), followed by Ajka Centre (42.5 µg kg^{-1}), Nagyvázsony (55.8 µg kg^{-1}), Eplény (70.6 µg kg^{-1}), the highest concentrations were observed in sample collected in Veszprém near the Bus station (768 µg kg^{-1}). In the background site, all PAHs were under the detection limit.

Table 11. Concentrations of PAHs individual in different villages (abbreviations, number of rings and molecular weight). LOQ: Limit of quantification

PAHs	No. of rings	Ajka Power plant [µg kg^{-1}]	Ajka Centre [µg kg^{-1}]	Veszprém Petrol station [µg kg^{-1}]	Veszprém Central bus station [µg kg^{-1}]	Nagy-vázsony [µg kg^{-1}]	Eplény [µg kg^{-1}]	Mean (range) [µg kg^{-1}]	Detection rate
Naphthalene	2	2.72	4.72	ND	1.93	4.6	2.22	2.7 (0-4.72)	83.3
2-methyl-naphthalene	2	2.01	2.58	1.04	2.21	2.52	1.76	2 (1.04-2.58)	100
1-methyl-naphthalene	2	1.15	2.64	1	2.44	1.93	1.05	1.7 (1-2.64)	100
Acenaphthylene	3	1	0.94	0.82	1.17	0.69	0.9	0.9 (0.69-1.17)	100
Acenaphthene	3	0.48	0.67	0.34	0.64	0.47	0.51	0.5 (0.34-0.67)	100
Fluorene	3	1.16	1.41	0.76	1.31	2.01	0.76	1.2 (0.76-2.01)	100
Phenanthrene	3	6.28	6.89	3.14	8.9	8.48	7.33	6.8 (3.14-8.9)	100
Anthracene	3	0.22	0.48	0.3	2.03	0.41	0.52	0.6 (0.22-2.03)	100
Fluoranthene	4	2.26	3.36	2.2	38.4	2.03	10.3	9.76 (2.03-38.4)	100
Pyrene	4	1.97	3.47	2.58	72.3	2.43	7.89	15.1 (1.97-72.3)	100
Benzanthracene	4	0.7	1.08	1.05	65.5	2.19	3.38	12.3 (0.7-65.5)	100
Chrysene	4	1.22	1.04	1.06	49.7	1.84	3.28	9.6 (1.04-49.7)	100
Benzo(b)fluoranthene	5	4.23	3.62	3.15	151	5.89	8.54	29.4 (3.15-151)	100
Benzo(k)fluoranthene	5	2.81	2.01	1.62	44	2.24	3.24	9.3 (1.62-44)	100
Benzo(e)pyrene	5	2.2	2.27	1.52	84.4	2.47	3.58	16 (1.52-84.4)	100
Benzo(a)pyrene	5	3.65	2.66	6.43	88.2	7.7	6.91	19.2 (2.66-88.2)	100
Dibenzo[a.h]anthracene	5	0.47	<LOQ	0.97	16.1	<LOQ	1.06	4.6 (0.47-16.1)	66.7
Indeno1.2.3CD-Pyrene	6	2.02	1.51	3.68	51.4	4.54	4.07	11.2 (1.51-51.4)	100
Benzo(g.h.i)perylene	6	2.07	1.23	3.09	85.6	3.41	3.36	16.4 (1.23-85.6)	100
Total PAHs		38.6	42.5	34.7	768	55.8	70.6		

Compared with the other sampling sites, concentration of HMW PAHs accumulated in *P. lanceolata* was significantly higher in Veszprém central bus station, showing relatively high level of pollution (Figure 15). According to our data, the most abundant individual PAHs accumulated in *P. lanceolata* were Phe and BbF in all sampling sites except Veszprém petrol station and Eplény, where BaP and Flu were detected in highest concentration (Table 12).

Concentration of Phe (3-ring) was 8.48 μg kg^{-1} dry-wt in Nagyvázsony, 6.89 μg kg^{-1} dry-wt in Ajka Centre, and 6.28 μg kg^{-1} dry-wt in Ajka power plant, while concentration of BbF (5-ring) from Veszprém central bus station, *Plantago* treated and Ajka power plant were 151 μg kg^{-1} dry-wt, 15.7 μg kg^{-1} dry-wt and 10.4 μg kg^{-1} dry-wt.

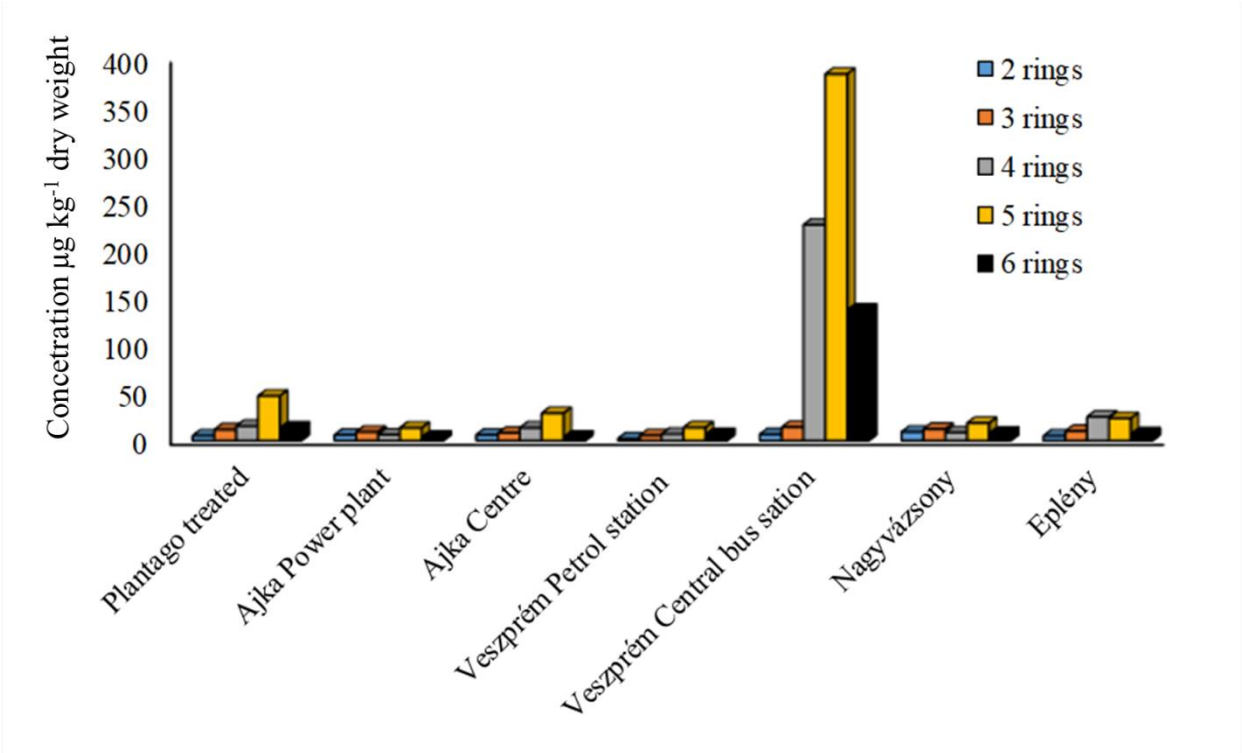

Figure 15. Total amount of different molecular weight PAHs on the sampling spots in the test plants

4. 4. 3. Comparison of experimentally treated plants and field collections

Strong correlation was found between the distribution pattern of accumulated PAHs in the experimentally treated sample with the collected *Plantago* samples (Table 12), with the only exception for Ajka Centre. Also, there were strong correlations between the different sites, except for Ajka Centre/Veszprém petrol station and Nagyvázsony/Veszprém bus station.

Table 12. Correlation between the collected Plantago samples

	Ajka Centre	Ajka power plant	Eplény	Nagyvázsony	*Plantago* treated	Veszprém petrol station	Veszprém central bus station
Ajka Centre	-	t=6.9656 df=17; $p=2.28*10^{-6}$; cor=0.8605	t = 3.7369; df = 17; p = 0.0016; cor= 0.6716	t = 8.8102; df = 17; $p= 9.582*10^{-8}$; cor= 0.9057	t=4.524; df=17; p=0.0003; cor=0.7391	t = 3.1421; df = 17; p = 0.0059; cor= 0.6061	t = 1.5791; df = 17; p = 0.1327; cor= 0.3577
Ajka power plant		-	t = 3.1818; df = 17; p = 0.00545; cor= 0.6109	t = 4.6852; df = 17; p = 0.0002128 cor= 0.7507	t = 1.8749; df = 17; p = 0.0781; cor= 0.4139	t = 1.3347; df = 17; p = 0.1996; cor= 0.3080	t = 0.36962; df = 17; p = 0.7162; cor= 0.0893
Eplény			-	t = 3.0815; df = 17; p = 0.006767; cor= 0.5987	t = 3.8569; df = 17; p = 0.001265; cor= 0.6831	t = 3.6883; df = 17; p= 0.00182; cor= 0.6667	t = 3.2482; df = 17; p = 0.0047; cor= 0.6188
Nagyvázsony				-	t = 5.0545; df = 17; $p =9.778*10^{-5}$; cor= 0.7749	t = 4.5884; df = 17; p= 0.000261; cor= 0.7438	t = 1.8438; df = 17; p = 0.0827; cor= 0.4082
Plantago treated					-	t = 5.6699; df = 17; $p=2.768*10^{-5}$; cor= 0.8088	t = 5.9182; df = 17; p =0.0000168; cor= 0.8205
Veszprém petrol station	-	-	-	-	-	-	t = 3.2381; df = 17; p = 0.004834; cor= 0.6176462
Veszprém central bus station	-	-	-	-	-	-	-

Both in the field and the experimentally treated *Plantago* samples, a high ratio of HMV PAHs was measured (Figure 16). The results showed that 5-ring PAHs represented 50% of total PAHs in Veszprém bus station and Plantago treated, followed by Veszprém petrol station

(39.5%), Ajka Centre (34.6%), Eplény centre (33%), Nagyvázsony roadside (32.8%) and Ajka power plant (24.8%) respectively. 6-ring PAHs where present in all samples and accounted for 6% to 20% of total PAHs.

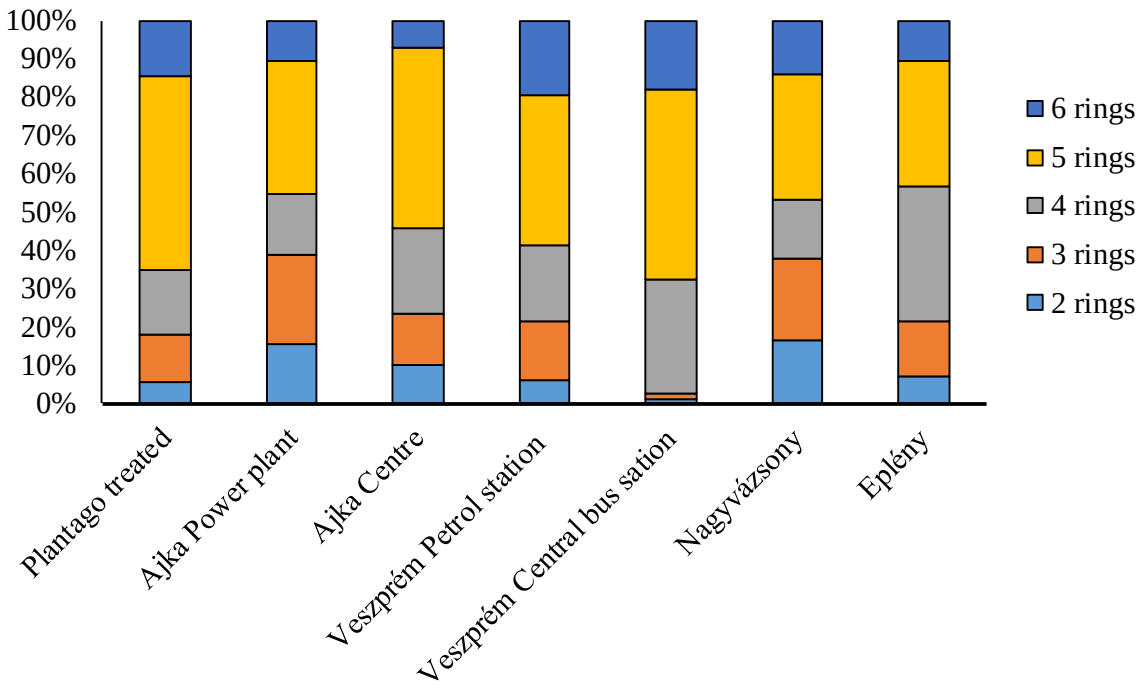

Figure 16. Percentage contribution of different molecular weight PAHs on the sampling spots in the test plants

These findings show that *Plantago* exhibits different bioaccumulation behaviour than other test species used in our previous studies. In these experiments lettuce (*Lactuca sativa* L.) was used as test species and was treated under laboratory conditions with urban aerosol extract. In this case lower molecular weight (LMW) PAH compounds were predominant in leaves and Nap and Ant had the highest BCF (Teke et al., 2020). These findings are consistent with data obtained in other field studies (e.g. An et al. 2017; Wang et al. 2017; Jia et al. 2018).

Nevertheless, accumulation pattern might highly depend on the taxon in question: A study by Huang et al. (2018) found higher accumulating tendency for light and medium molecular weight PAHs in oak leaves in contrary to mosses where stronger accumulating tendency for heavy molecular weight PAHs was measured. A study of Borgulat and Staszewski (2018) found relatively high share of 5- and 6-ring PAHs in grass. The study of Ashraf and Salam (2012) measured higher concentrations of DahA and B(g.h.i.)P in sampled vegetables such as cabbage. A transplanted pot study by Bakker et al. (1999) also detected the abundance of heavy molecular weight PAHs using *Plantago* test plants.

4. 4. 4. Comparison of sampling sites

In order to reduce the number of variables to two principal components (PC1 and PC2) and to establish the relationship between 19 PAHs in *Plantago* samples a Principal component analysis (PCA) was used. The biplot of PCA is presented in Figure 17. The PC1 component accounted for 68.09% of the total variance and the PC2 component accounted for 24.61% of the total variance. The first principal component (PC1) is in general associated with these 12 PAHs: Ant, Flt, Pyr, B(a)a, Cry, B(b)f, B(k)f, B(e)p, B(a)p, D(a,h)a, Ind, B(g,h,I,)p while 7 components: Nap, Flu, Me-Nap, Methy-Nap, Act, Ace, Phe from Σ19PAHs correlated with PC2.

The main sources of PAHs correlating with PC1 are markers of coal combustion (Pyr, BaA, and Cry) and markers of vehicle (gasoline and diesel) emissions (B(b)f, B(k)f and B(a)p,) or coal combustion and vehicle emission markers (Flt, Ant) (Limu et al., 2013). The PAHs correlating with PC2 (Acl, Ace, Flu, Phe, Ant) suggest substantial contribution from low temperature pyrolysis processes (like biomass combustion or coal combustion) (Yadav et al., 2020).

Two sites are clearly different: Pécsely National Park which is in fact served as a background sampling site and Veszprém Central bus station. In this station high environmental pollution is caused by the dominance of the relatively old diesel-powered, buses belonging to the Euro0–Euro3 European emission standards (Kovats et al., 2013).

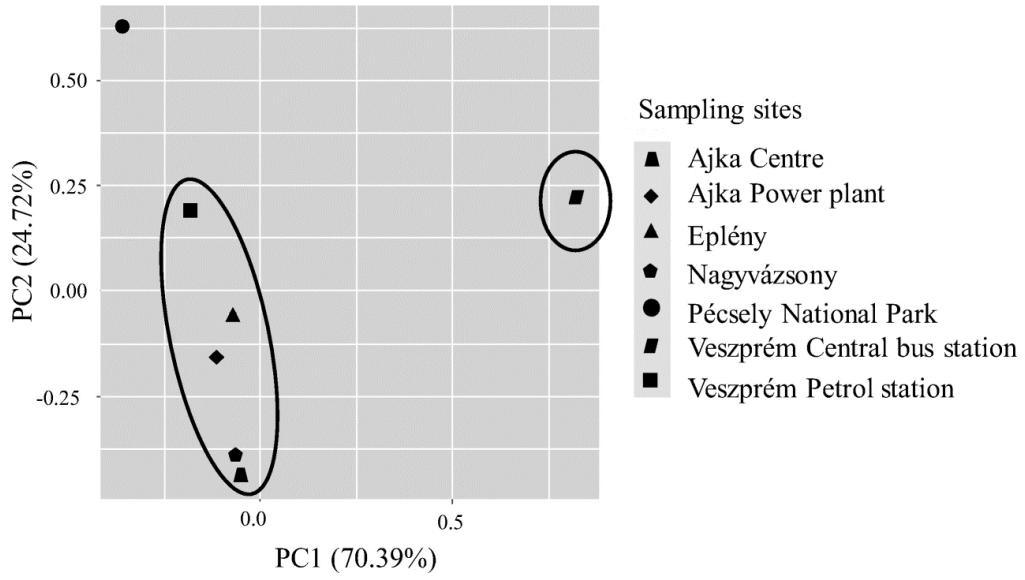

Figure 17. Principal component analysis (PCA) biplot based on the detected PAH concentrations from the sampling spots in the test plants

4. 4. 5. Source identification of PAHs in different villages

PAHs congener ratios have been widely used to distinguish between potential sources of PAHs in a number of previous works (reviewed by e.g. Tobiszewski and Namiesnik, 2012). 3 characteristic ratios have been selected to discriminate our samples. A Flt/(Flt+Pyr) ratio lower than 0.4 indicates petroleum, between 0.4 and 0.5 implies liquid fossil fuel combustion and over 0.5 implies combustion of grass, wood and coal (Tobiszewski and Namiesnik, 2012). In the case of Ind/(Ind + B(g,h,i)P), ratio between 0.2–0.5 indicates petroleum combustion, while value higher than 0.5 indicates biomass combustion (Yunker et al., 2002). B(a)a/(B(a)a+Cry) ratio over 0.35 has been defined to indicate combustion of vegetation and fossil fuel (Yunker et al., 2002).

In order to compare accumulation pattern in Plantago samples, B(a)a/(B(a)a+Cry) ratio against Flt/(Flt+Pyr) ratio as well as Ind/(Ind+BghiP) ratio against Flt/(Flt+Pyr) ratio were plotted (Figure 18; 19). Both plots indicate the difference between Ajka 1 and Veszprém 2 where biomass combustion seemed an important input. Contribution of biomass burning is obvious in case of Ajka power plant, as it uses app. 192.000 tons of wood/ year (Gyulai 2006). However, it is interesting to note that in addition to traffic, biomass combustion still provides a significant source despite the fact that sampling was done in June, well after the heating season.

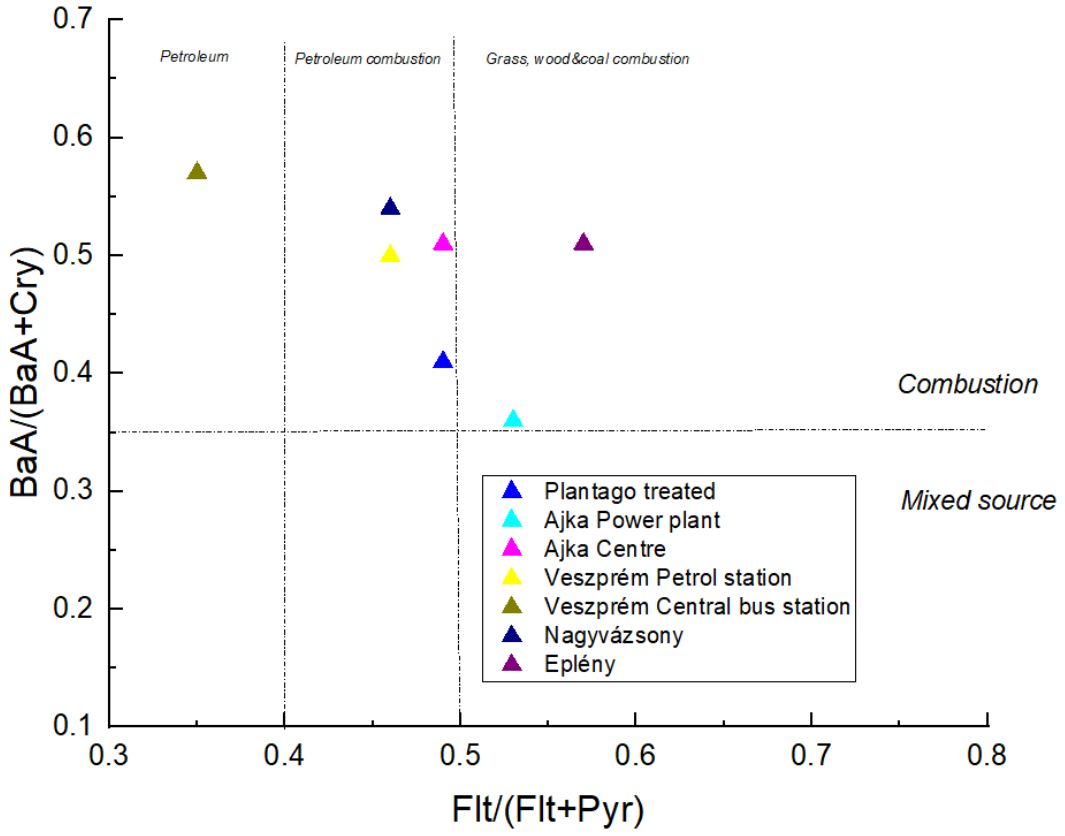

Figure 18. Cross plot of B(a)a/(B(a)a+Cry) ratio against Flt/(Flt+Pyr) for Plantago samples collected in the field

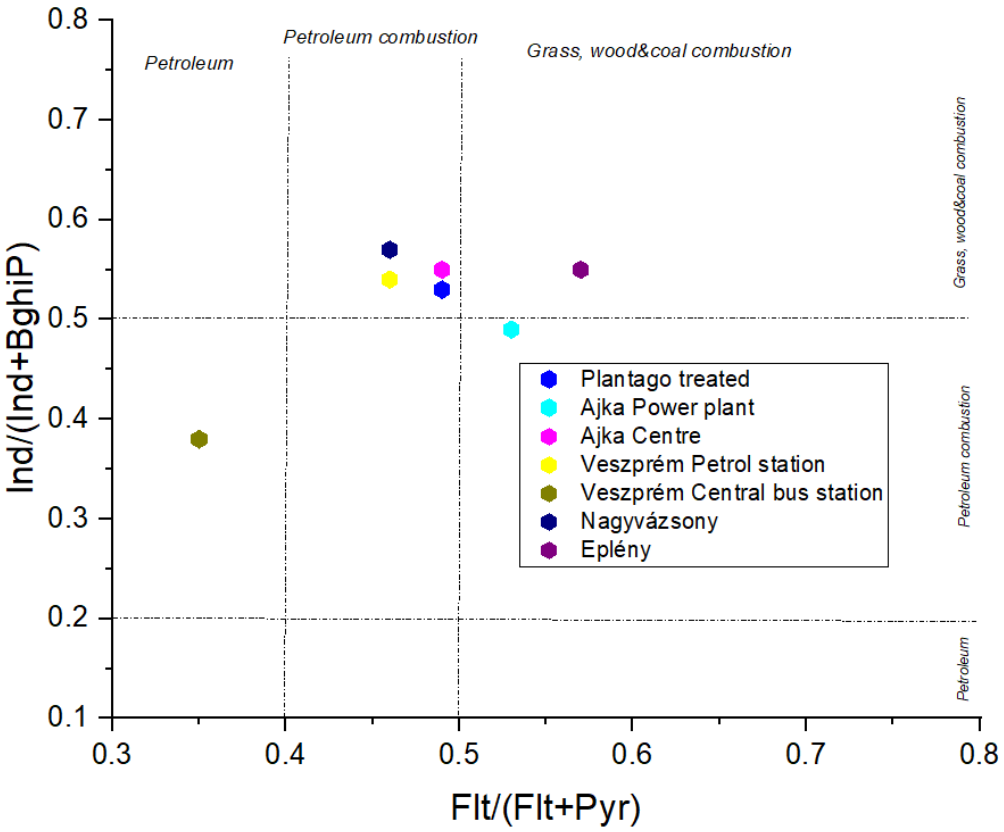

Figure 19. Cross plot of Ind/Ind+BghiP) ratio against Flt/(Flt+Pyr) for Plantago samples collected in the field

4. 5. Biomonitoring PAH levels in domestic kitchens by *Chlorophytum comosum* pot study

4. 5. 1. Assessing the PAH concentrations in the selected households

In the sampling site of HH1 the total PAHs concentration showed a clear time-dependency, being 127 µg kg^{-1} at the end of the first month of the exposure and 236 µg kg^{-1} at the end of the second month. This accumulation pattern was dominated by the low molecular weight PAHs, for example the concentration of Acenaphthylene was 1.2 µg kg^{-1} and 3.3 µg kg^{-1} ; Acenaphthene was 1 µg kg-1 and 1.7 µg kg^{-1} ; Fluorene 3.2 µg kg-1 and 5.7 µg kg^{-1} . However, some of the high molecular weight PAHs like Benzo[g,h,i]perylene showed a noticeable increase in the second month of the exposure, as its concentration was 1.1 µg kg^{-1} at the end of the first month but 8.1 µg kg^{-1} at the end of the second month. Concentration of the carcinogenic Benzo[a]pyrene was below 0 µg kg^{-1} at the first month of the exposure period but increased to 5.1 µg kg^{-1} by the end of the second month (Figure 20).

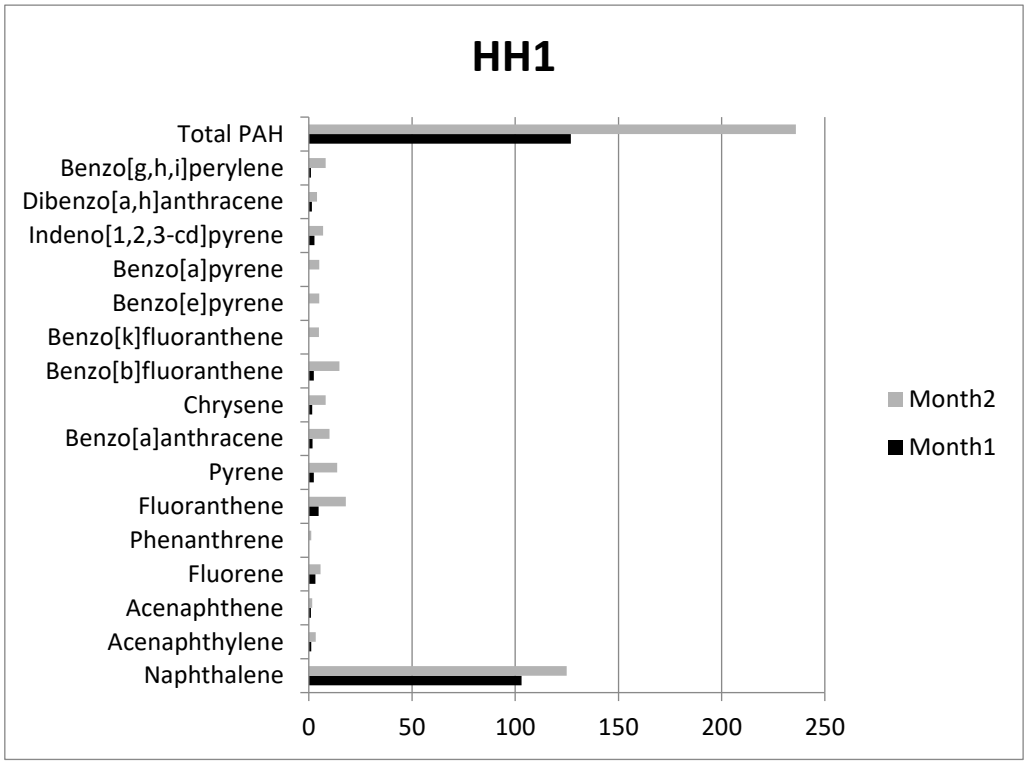

Figure 20. Accumulated PAHs (µg kg^{-1}) in Household1 after the first and second month

The dominant PAH was Naphthalene in all of the households, (HH1: 103 μg kg^{-1} and 125 μg kg^{-1}, HH2: 44 μg kg^{-1} and 260 μg kg^{-1}, in HH3: 195 μg kg^{-1}, each), which was similar to other studies (e.g. Zhu and Wang 2003, Sharma and Jain 2020). A study from China reported that 2- and 3-ring PAHs, especially Naphthalene, were dominant in household kitchens, while mainly 3- and 4-ring PAHs where dominant in commercial kitchens (Zhu and Wang 2003). However the production of Naphthalene might not be influenced by cooking styles according to a study by Huang et al., (2021). They compared volatile organic compounds emission in frying, steaming and grilling commercial kitchens and found that the three different cooking styles had similar production of Naphthalene.

In the sampling site of HH2, the total concentration of PAHs was 133.6 μg kg^{-1} and by the end of the second month the accumulated concentration showed a remarkable increase, reaching as much as 411.5 μg kg^{-1}. The biggest differences when comparing it to HH1 were the lack of certain PAH congeners: for example the carcinogenic Benzo[a]pyrene could not be detected, and the absence of No 6-ring PAHs was also a big difference.

There has been a dramatic increase in HH2 between Month1 and Month 2 which can be attributed to Naphthalene: its concentration increased from 44 to 260 μg kg^{-1}. A noticeable increase was also seen in the concentration of 3-ring PAHs, in case of Phenanthrene for example this increase was from 16 to 51 μg kg^{-1}. However the higher MW PAHs concentration showed a less clear tendency; for example concentration of the 5-ring Benzo[e]pyrene increased from 1 to 3.3 μg kg^{-1} while concentration of Benzo[k]fluoranthene remained practically the same (4.3 and 4.4 μg kg^{-1}) (Figure 21).

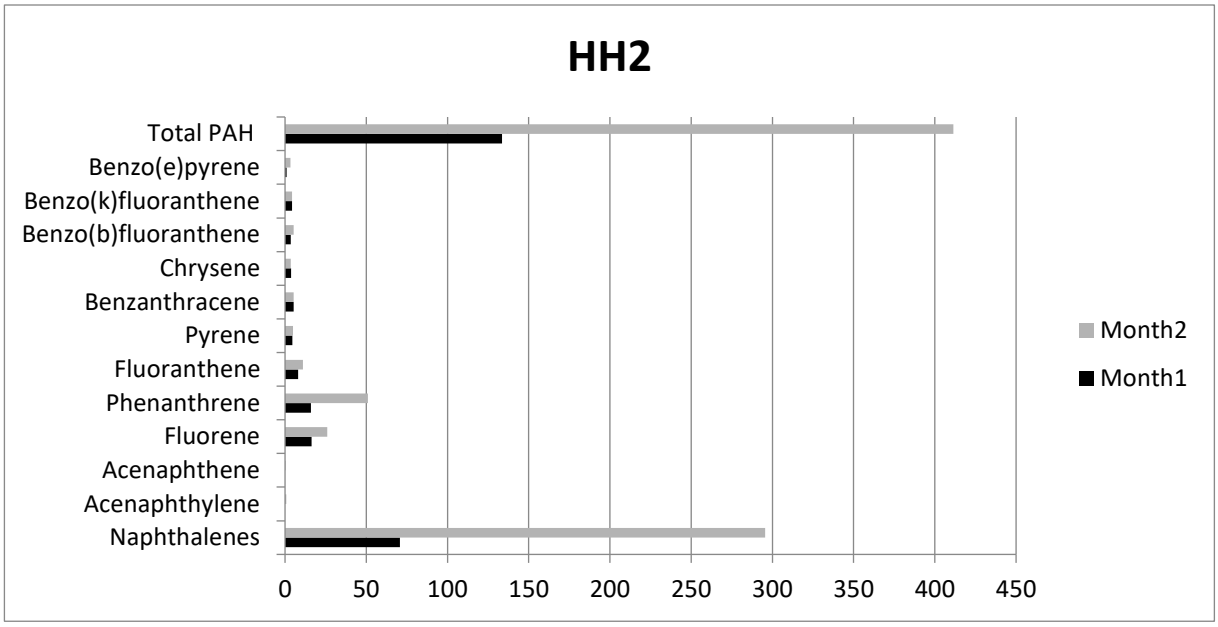

Figure 21. Accumulated PAHs (μg kg^{-1}) in Household 2 after the first and second month

In the sampling site of HH3 the results are only for the first month's exposure, due to the damage of the test system (Figure 22), though the total concentration of accumulated PAHs reached as much as 471.18 μg kg^{-1}. The second dominant PAH was Phenanthrene (124 μg kg^{-1}). Concentration of the carcinogenic Benzo[a]pyrene was also relatively high, 6.7 μg kg^{-1}.

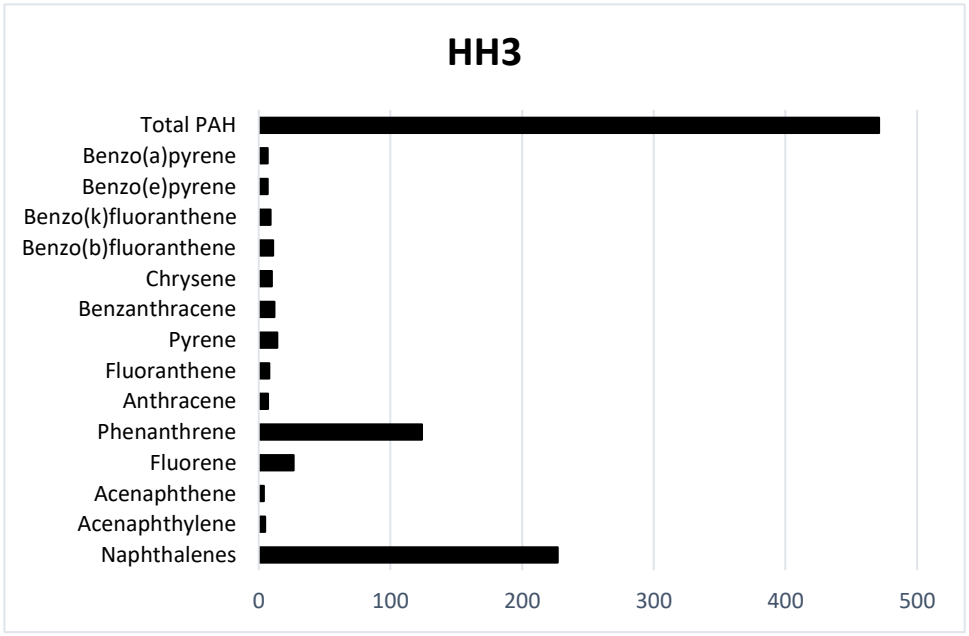

Figure 22. Accumulated PAHs (μg kg^{-1}) in Household3 after the first month

In the sampling site of HH4 (Figure 23) total amount of accumulated PAHs showed lower values than in the other sampling sites, 146 and 224.5 μg kg^{-1} by the end of the two exposure periods. The second dominant PAH similarly to the HH3 sampling site was Phenanthrene (37 and 45 μg kg^{-1}, respectively). Rather similar concentrations of the carcinogenic Benzo[a]pyrene were found when comparing Month1 and Month2, 1.9 and 2.3 μg kg^{-1}, respectively. However the concentration of Benzo[e]pyrene increased from 1.1 to 8.3 μg kg^{-1} by the end of the study.

Sun et al. (2020) found high concentrations of Phenanthrene when different Chinese cooking styles were compared. Phenanthrene was characteristic of Sichuan cuisine, which is mixed with quick frying, high-temperature cooking and large oil consumption. Phenanthrene was one of the most characteristic PAHs in the emissions from water-based cooking activities in residential Chinese kitchens (Zhao et al. 2019). In general, the relatively high share of Phenanthrene in HH2-HH4 sight might raise some health concerns because a study of Shin et al. (2013) found that in developed countries the main exposure pathway to Naphthalene and Phenanthrene was from the inhalation from indoor.

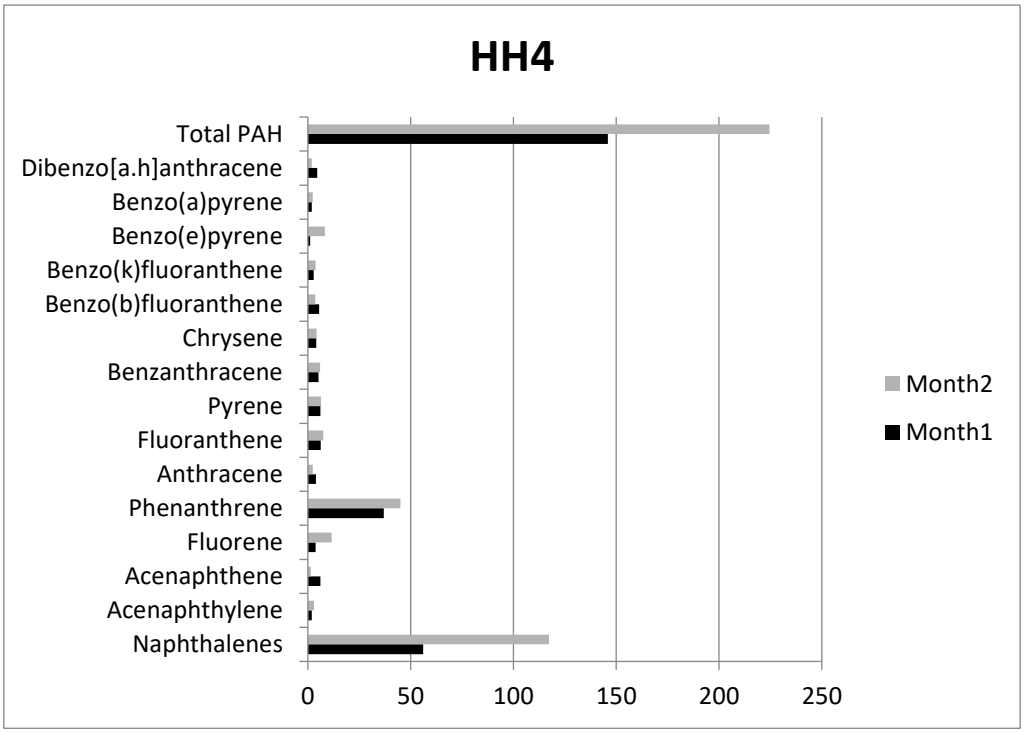

Figure 23. Accumulated PAHs (μg kg-1) in Household 4 after the first and second month

A study by See et al. (2006) reported that as a general rule, frying operations such as stir- and deep-frying generated higher molecular weight PAHs. These cooking methods are very common of Asian cooking: high amount of Indeno[1,2,3-cd]pyrene was found by Singh et al.

(2016) in samples taken in a North Indian commercial kitchen. Zhang et al. (2017b) also measured high concentrations of Benzo[a]pyrene, Benzo[b]fluoranthene, Benzo[a]anthracene, Indeno[1,2,3-cd]pyrene and Chrysene during domestic Chinese cooking. This might serve as an explanation for the relatively high share of these PAHs in HH1, as deep-frying accounts for app. 30 % of cooking operations.

High molecular weight PAHs were also found in university canteens and a charcoal-grilled chicken restaurant in Portugal (Vicente et al. 2021). Alves et al. (2021) compared emission of different size particulate matter (PM_{10}, $PM_{2.5}$ and PM_{1}) and total volatile organic compounds (TVOCs) during cooking different typical Latin meals such as stuffed chicken, fried mackerel, fried and grilled pork. They found that in general, the emissions from grilled pork contained PAHs in the highest concentration, including HMW PAHs such as Indeno[1,2,3-cd]pyrene, Dibenzo[a,h]anthracene and Benzo[g,h,i]perylene.

Hu et al. (2021) studied the behaviour of PAHs during deep frying with special regard to the mutagenic Benzo[a]pyrene and found that PAH level in sunflower oil generally raised with increasing frying time. It might serve as an explanation for the drastic increase of this PAH in HH1. In a study of Yao et al. (2015), Benzo[a]pyrene emission was found characteristic during deep frying. In our study, it was detected in households using this cooking method such as HH1, HH3 and HH4.

HH1 sampling site was also the only household where olive oil was used at relatively high frequency (app. 5%). In a study Wang et al. (2018c) compared particle emission characteristics originated from cooking with four types of oil (soybean oil, olive oil, peanut oil and lard) and also found that olive oil emitted the highest number of particles.

4. 5. 2. Accumulation capacity of Chlorophytum comosum

In the scientific literature numerus studies have reported the rather high accumulation rate of low molecular weight PAHs (Jia et al. 2018, Wang et al. 2017). Exposure pathways do differ to some extent: more volatile LMW PAHs are available in gas phase, while HMW PAHs are less volatile and occur mostly in particulates (Mukhopadhyay et al. 2020).

Furthermore, it is possible that the accumulation pattern highly depends on the taxon used (Huang et al., 2018). Capozzi et al. (2017) found that *Robinia pseudoacacia* leaves were able to accumulate both Low molecular weight and High molecular weight PAHs in a field study. Similar bioaccumulation capacity was reported for the perennial *Plantago lanceolata* (Bakker

et al. 1999, Hubai et al. 2021). Relatively high share of 5- and 6-ring PAHs was found in Poaceae species such as rice (Tao et al., 2006) and grass (Borgulat and Staszewski 2018). Positive correlation was found between atmospheric and accumulated PAH concentrations in maize (Lin et al., 2007).

Figure 25 shows the amount of PAH isomers in the different *Chlorophytum* samples. The relatively high share of 5- and 6-ring PAHs in HH1 at the end of the total exposure (33.9 µg kg^{-1} and 15.1 µg kg^{-1}) clearly shows that tis plant is capable to accumulate High molecular weight PAHs. 5-ring PAHs also reach remarkable concentrations in *Chlorophytum* leaves in HH2 and HH4 (13 µg kg^{-1} and 19.8 µg kg^{-1} by the end of the second month, respectively) and 33.6 in HH3 by the end of the first month of exposure.

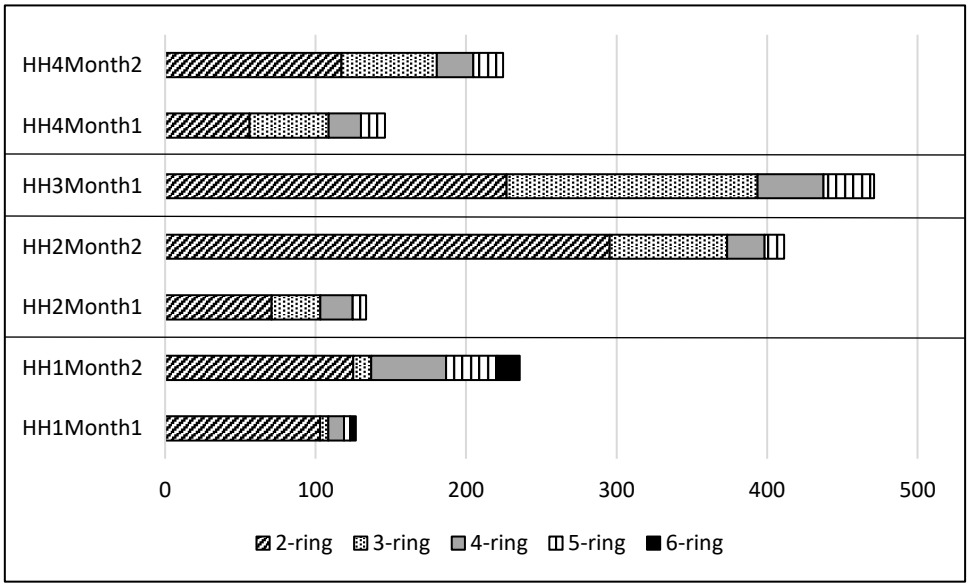

Figure 24. PAH isomers in Chlorophytum samples in HH1-HH4

4. 6. Evaluation of the 227 OECD Guideline for bioaccumulation studies

While bioaccumulation or biomonitoring studies have been extensively published using field collections, laboratory studies are rather rare in the literature. Lin et al. (2007) e.g. exposed roots and above-ground parts of maize plants to PAHs in air-tight bicameral exposure devices. The study established good correlations between accumulated compounds in parts of the plant tested and atmospheric PAHs' concentrations. Partially to fill this gap, computational/modelling studies are also available (e.g. Steyaert et al. 2009).

Lab-scale experimental studies are not only rare but very difficult to compare as different authors use very diverse approaches and equipment. These equipments are frequently 'home-made' such as the device mentioned above in Lin's study.

The experimental protocol defined in the 227 OECD Guideline provides every quality criteria: it specifies the conditions for the cultivation and treatment of plants, including the minimum number of test plants used in a treatment. The Guideline was adopted first by Kováts et al. (2017) to test the deleterious effects of PM-bound contaminants on higher plants. As the main aim of the study was to evaluate if the Guideline can be suitable to assess the magnitude and pattern of PAHs accumulation in laboratory-scale experiments, the benefits can be summarised as follows:

- Following a standardised protocol enhances reproducibility of the assays.
- Also, it increases comparability of different studies (for example, different atmospheric samples can be evaluated in successional test series).
- When comparing PAH accumulation pattern in experimentally treated plants and field collections, good correlations are achieved. The protocol provided by the Guideline creates adequate sensitivity.

5. Conclusions

Nowadays community gardening is more and more popular in every year. As such, the risk of the bioaccumulation of atmospheric polyaromatic hydrocarbons (PAHs) in vegetables grown in polluted areas can not be neglected. With some modifications the No. 227 OECD GUIDELINE the foliar uptake of PAHs from aqueous extract of an urban aerosol was proven suitable to test the deleterious effects of the water-soluble components of airborne particulate matter. Under laboratory conditions where the composition of the test materials is known, accumulated materials can be clearly correlated with the composition of the sample and the behaviour of different PAHs and pattern in bioaccumulation can be clearly followed.

The main benefit of such a standardised test is that concentration-effect relationships can be established. Under controlled conditions, with known pre-set exposure time; known composition of the sample the bioaccumulation capacity of different leafy vegetables could be measured with standard protocol. In this aspect, fixed exposure can be the most serious limitation. The Guideline determines the exposure of 21 or 28 days. Working with different species, it should be realised that they might have different growth rate. However, the OECD protocol strictly describes what age/stage the treatment should start. With the use of Guideline accumulation investigations can be planned with a high level of variability in timing of treatments or comparison between different species, etc.,

However, apart from leaf morphology and physiological properties, relative growth rate may also be an important factor affecting sensitivity. During the investigation of bioaccumulation capacity of rocket and lettuce had very similar accumulation rate, but they differ in their growth rate. While rocket showed high bioaccumulation potential, garden cress had one of the lowest ones. Striking differences were found in the accumulation potential of the vegetables which could be explained only partially with leaf morphological parameters. Most likely, different growth rates will also influence the concentration of accumulated PAHs. Accumulation pattern of the vegetables showed some similarity, with the predominancy of lower molecular weight (LMW) PAH compounds.

It can be concluded that using the protocol important information can be gathered on the accumulation pattern of each vegetable tested and clear dose-effect relationships can be drawn. Field studies are needed, however, to assess actual risk of consuming polluted vegetables.

Plants have the potential to improve urban air quality as leaves capture air pollutants e.g. PAHs from the air. The exposure of leaves to airborne PAHs is reflected in leaf PAH concentrations. Recent approaches (Zhu et al., 2008) attempt a quantitative relationship between air PAH level and plant PAH accumulation. In the pot study lettuce plants were used as passive monitors in small- and medium-sized villages. The accumulated PAHs concentrations in lettuce plants moved in a wide range. High concentrations were detected, these results were comparable to results measured in polluted areas in the world.

Biomass burning is an important source of PAHs. During the source appointment my data highlighted that PAHs are emitted during biomass burning, including household heating and burning of agricultural waste in the gardens as well. In addition, transport-related emissions also contribute. The fact that both the lettuce and soil samples showed the highest pollution in the Natura 2000 site Hárskút draws our attention to how important it is to analyze individual pollution sources.

In field collected Plantago samples the distribution of accumulated PAH isomers showed strong correlation with the experimentally treated sample. The study has shown that P. *lanceolata* is a reliable passive monitor when distribution pattern of PAH contamination is to be assessed. Every sample was characterised by the prevalence of HMW PAHs, in contrary with most of the reported studies. Also, experimental treatment under laboratory conditions provided a comparable reference to field collected samples

The people all around Europe ~~people~~ spend most of their time (approximately 90%) indoors (González-Martín et al. 2021). This new situation raised the necessity to examine air quality and air pollution levels indoor and also the potential health impacts. One of the major contributor which could increase the level of indoor air pollution is cooking (Zhai and Albritton 2020). Accumulation of PAHs in *Chlorophytum comosum* was investigated in 4 typical Hungarian kitchens over 1- and 2-month-long exposures. The size of selected households were similar the significantly differences were cooking practices and materials but the most important difference was the use of sunflower oil and deep-frying as a cooking method. In that households where the amount of used sunflower oil was higher there the relative share of HMW PAHs was considerably higher than in that which used lard and butter. The carcinogenic B(a)p was measured in detectable amounts where deep-frying was the mostly used cooking practice.

A clear time-dependent PAH accumulation pattern was demonstrated. The concentrations were considerably higher at the end of the 2 months period than after 1-month exposure. *C. comosum* as test plant was able to accumulate also LMW and HMW PAHs, so it has proven a sensitive biomonitor for indoor PAH levels. Its use as biomonitor is further increased by the popularity and easy-to-cultivate nature of this ornamental plant.

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8. Thesis points

1. I investigated if the treatment protocol described by the No. 227 OECD GUIDELINE FOR THE TESTING OF CHEMICALS: Terrestrial Plant Test: Vegetative Vigour Test is applicable for lab-scale bioaccumulation studies. Lettuce (*Lactuca sativa*) test plants were cultivated according to test conditions defined by the Guideline and treated by aqueous extract of urban aerosol. Significant PAH accumulation was experienced, calculated bioconcentration factors showed strong correlation with molecular weight of PAHs. The standard protocol defined by the Guideline proved fit to assess bioaccumulation pattern under controlled laboratory conditions.

2. Leafy vegetables were cultivated according to the Guideline and treated with the water extract of PM emitted by a diesel-powered light duty vehicle. These vegetables showed remarkable differences in the ratio and pattern of accumulated PAHs. Leaf celery and lettuce showed the highest, while sweet basil and garden cress the lowest accumulation rates. As the latter two are also recommended for cultivation in so-called balcony (rooftop) gardens, in the proximity of road traffic, the low accumulation rate might indicate the relative safety of this practice.

3. Accumulation pattern of PAHs was compared in experimentally treated and field *Plantago lanceolata* samples. Distribution pattern of accumulated PAHs showed strong correlation between the experimentally treated sample and most of the field plantain samples. This indicates that results gained during the experimental treatment protocol can be extrapolated to real-world conditions.

4. The applicability of *Chlorophytum comosum* 'Variegata' (Thunb.) Jacques (Family Asparagaceae, spider plant, syn. spider ivy, ribbon plant, or hen and chickens) was established in a pot study when PAH emission rates and patterns were monitored in previously selected rural kitchens. Distribution of PAHs in the biomonitor plants could be very clearly explained by the differences in the cooking styles and material usage in these households. Test plants were also able to accumulate both LMW and HMW PAHs. To my best knowledge, this was the first study which used *C. comosum* for indoor PAH bioaccumulation measurements.

5. In contrary to most of the reported literature, the accumulation of HMW PAHs was significant in a certain part of biomonitors investigated, especially in *P. lanceolata* and *C. comosum*.

6. Pattern of PAH accumulation in *Lactuca sativa* pot study showed a good discriminative power to assign different emission sources in small and medium-sized settlements taking part in the study.

9. List of publications

9. 1. Papers related to the dissertation

Teke, G.; Hubai, K.; Diósi, D.; Kováts, N. (2020): Assessment of Foliar Uptake and Accumulation of Airborne Polyaromatic Hydrocarbons Under Laboratory Conditions. Bulletin of Environmental Contamination and Toxicology, DOI: 10.1007/s00128-020-02814-z; **impact faktor: 2.151; Q2**

Hubai, K.; Kováts, N.; Sainnokhoi, T. A.; **Teke, G.** (2021): Accumulation pattern of polycyclic aromatic hydrocarbons using *Plantago lanceolata* L. as passive biomonitor. Environmental Science and Pollution Research, DOI: 10.1007/s11356-021-16141-1; **impact faktor: 4.223; Q2**

Kováts, N.; Hubai, K.; Sainnokhoi, T. A.; **Teke, G.** (2021): Biomonitoring of polyaromatic hydrocarbon accumulation in rural gardens using lettuce plants. Journal of Soils and Sediments, 21, 106–117. DOI: 10.1007/s11368-020-02801-1; **impact faktor: 3.308; Q1**

Kováts, N.; Hubai, K.; Diósi, D.; Hoffer, A.; **Teke, G.** (2022): Foliar Uptake and Accumulation of Polycyclic Aromatic Hydrocarbons from Diesel Emissions. Polycyclic Aromatic Compounds, 42(9), 6124-6135. DOI: 0.1080/10406638.2021.1977347 **impact faktor: 1.89; Q3**

Hubai, K.;Eck-Varanka, B.; Kováts, N.; **Teke, G.** (2023): Pot study using Chlorophytum comosum plants to biomonitor PAH levels in domestic kitchens. Environmental Science and Pollution Research, 30, 51932–51941. DOI: 10.1007/s11356-023-25469-9; **impact faktor: 5.019; Q1**

9. 2. Other papers

Üveges, V.; Ács, A.; Bíró, R.; Drávecz, E.; Hajnal, É.; Hubai, K. E.; Kacsala, I.; Kovács, K.; Kováts, N.; Kucserka, T.; Lengyel, E.; Matulka, A.; Selmeczy, G. B.; Stenger-Kovács, Cs.; Szabó, B.; **Teke, G.**; Vass, M.; Padisák, J. (2011): A vörösiszap katasztrófa hatása a Torna-patak és a Marcal élővilágára, a regeneráció első időszaka. Economica, 4 (12), 95-139.

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Kováts, N.; Fábián, V. A.; Hubai, K.; Diósi, D.; Sainnokhoi, T. A.; Békéssy, Zs.; **Teke, G.**: (2020): Seasonal Differences in Rural Particulate Matter Ecotoxicity. Aerosol Science and Engineering, 4, 169–177 DOI: 10.1007/s41810-020-00063-5; **impakt faktor: -; Q2**

Hubai, K.; Kováts, N.; **Teke, G.** (2021): Effects of urban atmospheric particulate matter on higher plants using *Lycopersicon esculentum* as model species. SN Applied Sciences, 3, 770, DOI: 10.1007/s42452-021-04745-8; **impact faktor:-**

Hubai, K.; Székely, O.; **Teke, G.**; Kováts, N. (2021): Is essential oil production influenced by air pollution in *Ocimum basilicum* L.?, Biochemical Systematics and Ecology, 96, 104248, DOI: 10.1016/j.bse.2021.104248.; **impact faktor: 1.381; Q3**

Kováts, N.; Hubai, K.; Diósi, D.; Sainnokhoi, T. A.; Hoffer, A.; Tóth, Á.; **Teke, G.** (2021): Sensitivity of typical European roadside plants to atmospheric particulate matter. Ecological Indicators, 24, 107428, DOI: 10.1016/j.ecolind.2021.107428.; **impact faktor: 4.958; Q1**

Kováts, N.; Hubai, K.; Sainnokhoi, T. A.; Hoffer, A.; **Teke, G.** (2021): Ecotoxicity testing of airborne particulate matter—comparison of sample preparation techniques for the *Vibrio fischeri* assay. Environmental Geochemistry and Health, 216, DOI: 10.1007/s10653-021-00927-w; **impact faktor: 4.609; Q1**

Kováts, N.; Hubai, K.; Sainnokhoi, T-A. ; Eck-Varanka, B.; Hoffer, A.; Tóth, Á.; Kakasi, B.; **Teke, G.** (2022): Ecotoxic emissions generated by illegal burning of household waste. Chemosphere, 298, 134263. **impact faktor: 8.12; D1**

Sainnokhoi, T. A.; Kováts, N.; Gelencsér, A.; Hubai, K.; **Teke, G.**; Pelden, B.; Tserenchimed, T.; Erdenechimeg, Z.; Galsuren, J. (2022): Characteristics of particle-bound polycyclic aromatic hydrocarbons (PAHs) in indoor PM2.5 of households in the Southwest part of Ulaanbaatar capital, Mongolia. Environmental Monitoring and Assessment, 194, 665. **impact faktor: 3.307; Q2**

Hubai, K.; Kováts, N.; Sainnokhoi, T. A.; Eck-Varanka, B.; Hoffer, A.; Tóth, Á., **Teke, G.** (2022). Phytotoxicity of particulate matter from controlled burning of different plastic waste types. Bulletin of Environmental Contamination and Toxicology, 109(5), 852-858. **impact faktor: 2.616; Q2**

Casotti Rienda, I.; Alves, C. A.; Nunes, T.; Soares, M.; Amato, F.; Sánchez de la Campa, A.; Kovats, N.; Hubai, K.; **Teke, G.** (2023): PM10 Resuspension of Road Dust in Different Types of Parking Lots: Emissions, Chemical Characterisation and Ecotoxicity. Atmosphere, 14(2), 305. **impact faktor: 3.222; Q2**

Hubai, K.; Kováts, N.; Eck-Varanka, B.; **Teke, G.** (2023): Pot study using Chlorophytum comosum plants to biomonitor PAH levels in domestic kitchens. Environmental Science and Pollution Research, 30(18), 51932-51941. **impact faktor: 5.053 ; Q1**

Kováts, N.; Hubai, K.; Sainnokhoi, T. A.; Eck-Varanka, B.; Hoffer, A.; Tóth, Á.; **Teke, G.** (2023): Ecotoxicity of PM10 emissions generated during controlled burning of waste PET. Environmental Toxicology and Pharmacology, 99, 104118. **impact faktor: 5.785; Q1**

9. 3. Congress attendances

Hajnal, É.; **Teke. G.**; Stenger-Kovács. Cs.; Padisák. J. (2010): Information in the biological datasets and the biodiversity estimation on the basis of the Peridat on-line database. AIS 2010, 5th International Symposium on Applied Informatics and Related Areas. 12.11.2010., Székesfehérvár, Hungary. **oral presentation**

Hubai, K. E.; Horváth, E.; Eck-Varanka, B.; **Teke, G.;** Tóth, Á.; Kováts, N.: Adapting and step by step refinement of the Vegetative Vigour Terrestrial Plant Test for assessing ecotoxicity of aerosol samples. 6th International Work-Conference on Bioinformatics and Biomedical Engineering, 25-27.04.2018. Granada, Spain. **poster presentation**

Kováts, N.; Hubai, K.; Gácsi, E.; Diósi, D.; **Teke, G.**; Tóth, Á.: Lab-scale tests for assessing bioaccumulation of atmospheric PM. Környezettoxikológiai Munkabizottság előadóülés, 30.05.2018. Veszprém, Magyarország, **oral presentation**

Kováts, N.; Diósi, D.; Hubai, K.; **Teke, G.**: Assessment os foliar uptake of polyaromatic hydrocarbons under laboratory conditions 7th Iberian Meeting Aerosol Science and Technology, 9-12.07.2019. Lisbon, Portugal. **poster presentation**

Hubai, K.; Békéssy, Zs.; Kováts, N.; Diósi, D.; Gácsi, E.; **Teke, G.**; Paulovits, G.: Sensitivity of roadside plant community to particle bound pollutants. 7th Iberian Meeting Aerosol Science and Technology, 9-12.07.2019. Lisbon, Portugal. **poster presentation**

Teke, G.: Estimation of polyaromatic hydrocarbon bioaccumulation in rural kitchen gardens. VEAB Környezettoxikológiai Munkabizottsága 2020. évi I. Tudományos Ülés, 06.03.2020. Veszprém, Hungary. **oral presentation**

Eck-Varanka, B.; Hubai, K.; Horváth, E.; Kováts, N.; **Teke, G.;** Tóth, Á.: Assessing Ecotoxicity of Size-fractionated Airborne Particulate Matter. Central Asian DUst Conference (CADUC) 8-12.04.2019. Dushanbe, Tadjikistan. **oral presentation**

10. Supplementary material

1. Annex: Repeatability and Accuracy values of PAH analytes in spiked plant samples.

	measured spiked sample (μg kg^{-1})					average (μg kg^{-1})	SD	RSD	Assigned value (μg kg^{-1})	Recovery %
	1	2	3	4	5					
Naphthalene	31.00	21.12	29.00	21.15	26.00	25.65	4.49	17.51	25.00	102.6
2-methylnaphthalene	26.50	20.98	26.11	22.24	21.12	23.39	2.71	11.58	25.00	93.6
1-methylnaphthalene	30.11	25.00	20.24	22.26	25.25	24.57	3.72	15.15	25.00	98.3
Acenaphthylene	24.12	29.85	25.25	24.11	20.15	24.70	3.47	14.06	25.00	98.8
Acenaphthene	23.63	29.25	25.52	20.28	27.00	25.14	3.41	13.55	25.00	100.5
Fluorene	29.00	22.12	28.00	26.00	26.00	26.22	2.64	10.05	25.00	104.9
Phenanthrene	20.29	30.09	27.65	27.01	25.52	26.11	3.65	13.97	25.00	104.4
Anthracene	30.11	20.11	23.32	32.00	27.40	26.59	4.87	18.33	25.00	106.4
Fluoranthene	23.65	30.54	20.30	21.11	20.30	23.18	4.34	18.72	25.00	92.7
Pyrene	20.00	20.22	23.00	23.35	26.26	22.57	2.58	11.41	25.00	90.3
Benzo(a)anthracene	23.00	24.14	22.11	20.23	26.00	23.10	2.16	9.37	25.00	92.4
Chrysene	22.00	21.00	21.50	22.25	24.18	22.19	1.21	5.47	25.00	88.7
Benzo(b)fluoranthene	22.00	22.00	22.00	25.01	23.70	22.94	1.37	5.97	25.00	91.8
Benzo(k)fluoranthene	23.00	23.00	20.24	20.05	24.02	22.06	1.80	8.16	25.00	88.3
Benzo(e)pyrene	29.00	20.16	22.00	23.00	26.70	24.17	3.60	14.90	25.00	96.7
Benzo(a)pyrene	25.10	22.54	22.11	20.00	23.54	22.66	1.88	8.29	25.00	90.6
Indeno(1,2,3-cd)pyrene	26.00	25.00	23.82	27.59	21.00	24.68	2.48	10.05	25.00	98.7
Dibenzo(a,h)anthracene	21.55	20.61	20.45	24.54	26.62	22.75	2.72	11.93	25.00	91.0
Benzo(g,h,i)perylene	21.20	26.20	20.70	22.24	22.56	22.58	2.16	9.56	25.00	90.3

2. Annex: Repeatability and Accuracy values of PAH analytes in spiked aerosol filter water extract spiked samples

	measured spiked sample (mg l^{-1})					average ($\mu g\ l^{-1}$)	SD	RSD	Assigned value ($\mu g\ l^{-1}$)	Recovery %
	1	2	3	4	5					
Naphthalene	0.107	0.115	0.085	0.103	0.106	0.10	0.01	10.74	0.10	103.2
2-methylnaphthalene	0.119	0.103	0.102	0.103	0.105	0.11	0.01	6.70	0.10	106.4
1-methylnaphthalene	0.110	0.117	0.113	0.113	0.103	0.11	0.01	4.62	0.10	111.2
Acenaphthylene	0.104	0.110	0.115	0.111	0.104	0.11	0.00	4.52	0.10	108.8
Acenaphthene	0.103	0.104	0.093	0.100	0.110	0.10	0.01	6.04	0.10	101.9
Fluorene	0.108	0.103	0.104	0.111	0.118	0.11	0.01	5.59	0.10	108.7
Phenanthrene	0.102	0.085	0.102	0.087	0.092	0.09	0.01	8.71	0.10	93.7
Anthracene	0.102	0.108	0.115	0.114	0.117	0.11	0.01	5.46	0.10	111.1
Fluoranthene	0.117	0.104	0.106	0.103	0.106	0.11	0.01	5.23	0.10	107.3
Pyrene	0.095	0.104	0.106	0.102	0.106	0.10	0.00	4.30	0.10	102.4
Benzo(a)anthracene	0.119	0.086	0.100	0.086	0.108	0.10	0.01	14.35	0.10	99.8
Chrysene	0.104	0.104	0.094	0.093	0.104	0.10	0.01	5.77	0.10	99.8
Benzo(b)fluoranthene	0.103	0.102	0.112	0.103	0.105	0.11	0.00	3.82	0.10	105.0
Benzo(k)fluoranthene	0.106	0.115	0.110	0.103	0.104	0.11	0.00	4.54	0.10	107.6
Benzo(e)pyrene	0.102	0.104	0.103	0.098	0.105	0.10	0.00	2.46	0.10	102.2
Benzo(a)pyrene	0.081	0.109	0.097	0.099	0.110	0.10	0.01	11.81	0.10	99.2
Indeno(1,2,3-cd)pyrene	0.102	0.108	0.105	0.109	0.104	0.11	0.00	2.83	0.10	105.5
Dibenzo(a,h)anthracene	0.113	0.105	0.110	0.094	0.103	0.10	0.01	6.89	0.10	104.9
Benzo(g,h,i)perylene	0.103	0.105	0.103	0.099	0.103	0.10	0.00	2.11	0.10	102.5

3. Annex: LOD and LOQ values of PAH analytes in spiked plant samples.

	LOD (mg kg^{-1})	LOQ (mg kg^{-1})
Naphthalene	0.0100	0.031
2-methylnaphthalene	0.0120	0.036
1-methylnaphthalene	0.0110	0.033
Acenaphthylene	0.0105	0.032
Acenaphthene	0.0120	0.036
Fluorene	0.0110	0.033
Phenanthrene	0.0100	0.030
Anthracene	0.0110	0.033
Fluoranthene	0.0100	0.032
Pyrene	0.0110	0.033
Benzo(a)anthracene	0.0110	0.033
Chrysene	0.0130	0.039
Benzo(b)fluoranthene	0.0110	0.033
Benzo(k)fluoranthene	0.0110	0.033
Benzo(e)pyrene	0.0130	0.039
Benzo(a)pyrene	0.0125	0.038
Indeno(1,2,3-cd)pyrene	0.0140	0.042
Dibenzo(a,h)anthracene	0.0150	0.050
Benzo(g,h,i)perylene	0.0140	0.042

4. Annex: LOD and LOQ values of PAH analytes in aerosol filter water extract spiked samples.

	LOD (mg l^{-1})	LOQ (mg l^{-1})
Naphthalene	0.00027	0.00085
2-methylnaphthalene	0.00028	0.00089
1-methylnaphthalene	0.00027	0.00084
Acenaphthylene	0.00025	0.00094
Acenaphthene	0.00024	0.00092
Fluorene	0.00021	0.00082
Phenanthrene	0.00020	0.00078
Anthracene	0.00017	0.00063
Fluoranthene	0.00015	0.00059
Pyrene	0.00021	0.00078
Benzo(a)anthracene	0.00018	0.00069
Chrysene	0.00022	0.00085
Benzo(b)fluoranthene	0.00021	0.00079
Benzo(k)fluoranthene	0.00023	0.00089
Benzo(e)pyrene	0.00024	0.00092
Benzo(a)pyrene	0.00026	0.00099
Indeno(1,2,3-cd)pyrene	0.00030	0.00098
Dibenzo(a,h)anthracene	0.00028	0.00100
Benzo(g,h,i)perylene	0.00025	0.00100

5. Chromatogram of the accumulated PAHs in plant samples