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Particulate Matter Accumulation on Apples and Plums: Roads Do Not Represent the Greatest Threat

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Abstract: Particulate matter (PM) is a mixture of solid and liquid substances of organic and inorganic character suspended in air. Plants are used as biological filters of air. However, PM can be deposited on their edible parts, with a negative effect on people's health. The aim of this study was to document the PM accumulation on apples and plums harvested from orchards located alongside roads with differing amounts of traffic. Plums accumulated more PM than apples. The deposition of PM on apples increased during fruit development and was highest at harvest. The impact of road type, traffic intensity, and distance from the road on PM accumulation on fruit was small. The least PM was adsorbed by apples harvested from an orchard located close to a road with the highest traffic, while in the case of plums, no effect of the road on PM deposition was recorded. The amount of PM accumulated on fruits depended on the species (fruit morphology, harvest period), activities undertaken in the orchard (early pruning exposes fruits to PM, ecological preparations increase fruit viscosity), and sources of pollution other than the roads located close to the orchard. Washing fruits with water removed half of the accumulated PM.

Keywords: fruits; orchards; food safety; road type; air pollution; particulate matter

1. Introduction

Air pollution represents an increasing threat to human health and one of the most dangerous inhaled pollutants is particulate matter (PM) [1]. PM is a mixture of solid and liquid substances of organic and inorganic character suspended in air. It comes from a wide range of natural (forest fires, volcanic activity, dust storm) and anthropogenic (domestic heating, incomplete combustion of fuels in car engines, abrasion of roads and tyres, industrial or construction activities) sources [2]. The toxicity of PM is determined by its size, chemical composition, origin, solubility, ability to generate reactive oxygen, and reactivity in the air [1,3]. Particles < 10 μm in diameter (PM_{10}) after inhalation can invade the lungs and even reach the bloodstream [1]. $PM_{2.5}$ has a small diameter (<2.5 μm) and unlike the larger PM is capable of carrying toxic compounds that pass through the respiratory tract and damage other organs due to air/blood exchange in the lungs [1,3]. The contents of risk elements bound to PM particles vary depending on the site, source, traffic level, seasonal variability and meteorological parameters [4], and include numerous hazardous components such us toxic or carcinogenic trace elements [4,5], polycyclic aromatic hydrocarbons [4,6], and pesticides [7].

Urban plants, particularly trees and shrubs, are used as biological filters, with foliage passively purifying the ambient air [8]. Air purification by plants is one of the most important ecosystem services provided by greenery in urban and industrial areas [9]. However, edible crops grown in a polluted environment can be contaminated with various toxic compounds contained in PM, which can have a serious impact on food safety [5,10–14]. Furthermore, even if PM is of a relatively low concentration, there is a possibility that pollutants may accumulate in the edible parts of plants beyond safe levels if the exposure time is extended [13]. Relatively high concentrations of polycyclic aromatic hydrocarbons, which easily accumulate in the hydrophobic surface layers of plants [11,15], and trace elements, including heavy metals less bioavailable in soils [5,13,16–18], have been recorded in the edible parts of vegetables and fruits. Where this is the case, fruits in case of which high atmospheric deposition has been determined as the principal accumulation pathway of pollutants could be great source of harmful substances for humans, comparable to inhalation and dermal contact. According to Wang et al. [19], 6.34%, 76.5%, and 28.1% of apple peel samples exceeded China's maximum permissible levels for Cd, Cr, and Zn respectively. Furthermore, in some conditions, compared with PM sources trace elements bioaccessibility was significantly higher for plant surfaces [20], probably due to chemical speciation, toxicity and mobility changes, which can be explained by the activity of phyllosphere organisms capable of releasing inorganic and organic compounds possessing acidifying, chelating and/or reductive properties [21]. However, Grembecka and Szefer [22] have reported that pome fruits originating from different geographical areas are not rich in trace elements (Zn, Cu, Mn, Cr). Absorption of polycyclic aromatic hydrocarbons through the consumption of fruits and vegetables has been estimated to be between 5 and 10% on average, which is about 31% of the total food intake [23]. In Catalonia, daily intake of polycyclic aromatic hydrocarbons due to fruit consumption has been estimated to be 0.08–0.258 μg day^{-1} [23]. In China, the daily dietary polycyclic aromatic hydrocarbons intake via vegetable consumption is slightly higher than inhalation exposure of ambient atmospheric polycyclic aromatic hydrocarbons [10,24]. However, it should be borne in mind that significant disparities exist in the daily intake of polycyclic aromatic hydrocarbons (but most probably other pollutants as well) via ingestion of plant products because (i) fruits and vegetables may make up a large proportion of people's diets, and (ii) levels of polycyclic aromatic hydrocarbons may be very different depending on the crop's environment, storage or transformation processes [11].

It is worth noting that the adverse effects of airborne PM on edible plants (biomass reduction, mechanical damage) are not always apparent, suggesting that these may be accumulated in high amounts on the plant's surface and in wax before symptoms of toxicity become visible [25]. Similar observations have been made for cabbage and spinach treated with PM enriched with heavy metals. Despite the high foliar accumulation of pollutants, no toxicity symptoms have been recorded [20]. In cabbage leaves, the cuticular wax appears too thick for PM to induce any morphological changes at the leaf surface even if it is accumulated in large quantities [20]. For this reason alone, it is very difficult to distinguish a plant product that has accumulated airborne pollutants from a clean one. Finally, contaminates can cumulate in processed end products such as juices, jams or oils [26].

Diesel and petrol exhaust emissions together with non-exhaust road/vehicle sources (resuspension, brakes, tyres and road surface wear, motor lubricant oils) are important sources of PM pollution, including soot, organic particles, Fe-rich particles, S-rich particles, Mn-rich particles, and Ca-rich particles [27]. The consequence of road traffic is increased air concentrations of gaseous pollutants, polycyclic aromatic hydrocarbons, and trace elements [28–32]. Global PM emissions from on-road vehicles will increase in future to 1050 Gg in 2030 and to as much as 1260 Gg in 2050 [33]. In orchards, an important source of PM may be application of fertilisers, ripening agents and pesticides [7,34]. The aim of this study therefore was therefore to document the accumulation of PM by popular fruits (apples and plums) harvested from trees grown in three orchards located alongside roads carrying differing levels of traffic.

2. Materials and Methods

2.1. Experimental Locations

2.1.1. Apple Orchards

The experiment was carried out in three orchards located close to asphalt roads with different traffic intensity and structure, which were assessed in June (Table 1).

Table 1. Traffic intensity and structure on the roads closest to the apple orchards. Data are means ± SE, *n* = 3.

Parameter		Rural Location	Local Road	National Road
Apple Orchards (Vehicles·h^{-1})				
Traffic density	7.00–8.00	15.3 (±2.7) [1]	110.3 (±6.8)	395.0 (±13.2)
	12.30–13.30	9.1 (±2.2)	92.7 (±5.5)	374.2 (±16.7)
Traffic structure		Large share of agricultural vehicles (e.g., tractors) in the morning hours. Passenger cars of all ages, rarely over 10–15 years old.	All kinds of cars, including trucks and agricultural vehicles. Different age and condition of vehicles.	All kinds of cars, including trucks, except for agricultural vehicles. Different age and condition of vehicles.

[1] based on own calculations.

The first experimental site was an intensively managed orchard in the village of Wólka Łęczeszycka (51°45′59″ N, 20°46′43″ E) far away from local or national roads, denoted in this paper for simplicity as 'rural location'. In this orchard, all the necessary agrotechnical and chemical treatments were performed. During the experimental period, construction work was being carried out 100 m away from the orchard. The examined orchard and construction site were separated from each other by other orchards. Fruits were sampled from the first (80 m from the local road, distance A), third (82.5 m from the local road, distance B), and tenth rows of trees (92.5 m from the local road, distance C). The second experimental site was an orchard with integrated, extensive fruit production located in the village of Ignaców (51°45′33″ N 20°38′45″ E) close to a local, but busy road (high number of vehicles, low speeds), denoted in this paper for simplicity as 'local road'. In this orchard conventional cultivation practices were mixed with an ecological approach in accordance with the guidelines of integrated fruit production. This orchard comprised several apple varieties, which resulted in an increased frequency of runs by mechanical equipment in the orchard during treatments and harvest. Fruits were sampled from the first (11 m from the busy local road, distance A), third (13 m from the busy local road, distance B), and tenth rows of trees (23 m from the busy local road, distance C). The third experimental site was an intensively managed orchard located in the village of Wólka Łęczeszycka (51°45′59″ N, 20°46′43″ E) close to voivodship road no. 728 (very high number of vehicles, high speeds), denoted in this paper for simplicity as 'national road'. In this orchard all the necessary agrotechnical and chemical treatments were performed. Unlike the other locations, the road at this site was at a higher altitude than the crops growing next to it. Fruits were sampled from the first (17 m from the busy national road, distance A), third (19.5 m from the busy national road, distance B), and tenth rows of trees (29 m from the busy national road, distance C).

The approximate number of agrotechnical and chemical treatments in apple orchards is presented in Table 2. In every examined orchard similar number of treatments was performed, but in the Ignaców orchard (donated as 'local road') only treatments permitted in integrated fruit production were possible.

Table 2. Number of agrotechnical and chemical treatments in apple orchards.

Treatment	Plant Protection (Pests, Diseases)	Herbicides	Foliar Fertilization	Soil Fertilization	Chemical Thinning	Mechanical Pruning	Mowing	Harvest
	Number·row^{-1}							
	22–24	2–3	8–10	2	1–2	1	2–3	8–12

In apple orchards, the concentration of ambient PM was measured with The Dust Air Personal Controler (Central Mining Institute, Katowice, Poland). Measurements were carried out on the fifteenth day of each month (June–October) in three replicates. A single replication was a 3-min measurement. All the results obtained were low, the values were below the acceptable limits. Slightly higher PM_{10} and $PM_{2.5}$ concentrations were recorded in the rural location, most probably due to construction works which took place in June–July (Table 3).

Table 3. Concentrations of PM_{10}, $PM_{2.5}$ and PM_1 in the apple orchards. Data are means ± SE, n = 15.

Parameter	Rural Location	Local Road	National Road
		μg m^{-3}	
PM_{10}	17.5 (±1.07)	15.3 (±0.84)	15.2 (±0.25)
$PM_{2.5}$	14.8 (±1.37)	13.5 (±0.43)	13.2 (±0.33)
PM_1	11.5 (±0.31)	12.1 (±0.35)	11.7 (±0.30)

2.1.2. Plum Orchards

The three plum orchards examined in this study were located close to asphalt roads with different traffic intensity and structure, which were assessed in June (Table 4). All three were intensively managed with all the necessary agrotechnical and chemical treatments performed. The first experimental site was located in the village of Zakrzew (51°49′38″ N 20°28′33″ E). The plum trees were a long way from any type of road, denoted in this paper for simplicity as 'rural location'. Fruits were sampled from the first row of trees (150 m from the local road). The second experimental site was situated in the village of Olszew (51°46′14″ N, 20°49′33″ E). This orchard was adjacent to the local road (high number of vehicles, low speeds), denoted in this paper for simplicity as 'local road'. Fruits were sampled from the first row of trees (3.5 m from the local road). The third experimental site was situated in the village of Podgóra (51°22′34″ N, 21°26′31″ E). This orchard was located close to the national road no. 729 (high number of vehicles, high speeds), denoted in this paper for simplicity as 'national road' Fruits were sampled from the first row of trees (14 m from the road).

Table 4. Traffic intensity and structure on the roads closest to the plum orchards. Data are means ± SE, n = 3.

Parameter		Rural Location	Local Road	National Road
			Plum Orchards (Vehicles·h^{-1})	
Traffic density	7.00–8.00	72.0 (±12.1) [1]	78.0 (±15.7)	417.3 (±27.5)
	12.30–13.30	66.3 (±6.78)	70.3 (±10.0)	398.7 (±22.6)
Traffic structure		All kind of kind of vehicles, however large share of agricultural vehicles (e.g., tractors) was recorded in the morning hours. Passenger cars of all ages, rarely over 10–15 years old.	All kinds of cars, including trucks. Different age and condition of vehicles.	All kinds of cars, including trucks except for agricultural vehicles. Different age and condition of vehicles.

[1] based on own calculations.

The approximate number of agrotechnical and chemical treatments in plum orchards is presented in Table 5. In every examined orchard similar number of treatments was performed.

Table 5. Number of agrotechnical and chemical in plum orchards.

Treatment	Plant Protection (Pests, Diseases)	Herbicides	Foliar Fertilization	Soil Fertilization	Chemical Thinning	Mechanica lPruning	Mowing	Harvest
				Number·row^{-1}				
	4–6	3–4	2–4	1–2	-	1	4–5	4–5

2.2. Plant Material

The objects of this study were two fruit tree species: Malus domestica Borkh. (apple tree, cultivar 'Gloster') and Prunus domestica L. (plum tree, cultivar 'Cacańska Lepotica'). Plants had already been growing in vivo in the selected locations for several years and within the same species were approximately the same age and size in all three orchards (apples: 7 years, plums: 10 years). The tree rows faced nearby roads. The spacing between rows was 3.5 m (apples) or 4.5 m (plums) with 1.2 m (apples) or 2.5 m (plums) between trees in the row. The studied orchards were located close to one other (apples within 0.8–10 km, plums between 25 and 52 km), therefore the plants were growing in almost identical microclimatic conditions (rainfall, wind). The ground cover in each orchard was the same and thus had no impact on PM accumulation by fruits.

Apple fruits were sampled on two dates (July: 50–60% size of mature fruit/BBCH (Biologische Bundesanstalt, Bundessortenamt und CHemische Industrie)-scale 75–76 and October: harvest maturity/BBCH-scale 87–89), while plums were sampled once (July: harvest maturity/BBCH-scale 87–89). The fruits were sampled from three trees (biological replications) at three distances (apples) or one distance (plums) from the road, from the entire cross-section of the tree and from branches located 1.0–1.5 m above the ground. The harvested fruits were free from pests, disease, and mechanical damage. Each sample consisted of 12 apples (for analysis 3–4 fruits with a skin surface of approximately 400 cm^2 were randomly selected) or 32 plums (for analysis 8–12 fruits with a skin surface of approximately 400 cm^2 were randomly selected) and was stored in an individual plastic box for no longer than 48 h before analysis.

Accumulation of PM and wax content on the apples and plums were compared on fruits sampled from three orchards at harvest maturity (apples: October, plums: July) and from the same distance from the road (first row from the adjacent road).

2.3. Quantitative Assessment of PM and Fruit Wax Content

The amount of water-insoluble PM was analysed according to Dzierżanowski et al. [35]. Two categories of PM were determined: (i) water-washable from fruit surfaces ($_{S}$PM, in nature washed away by rain), and (ii) retained in fruit wax ($_{W}$PM). The fruits were first washed for 60 s with 250 mL distilled water and then for 30 s with 150 mL chloroform (≥99% stabilised). The volume and duration of chloroform treatments were determined before the analysis in order to collect accumulated PM most effectively without dissolving the top layers of the epidermis. The washing solutions were first sieved through a metal sieve (retention 100 µm, Haver & Boecker, Oelde, Germany) and then filtered through a 10 µm paper filter (Whatman, UK, Type 91), followed by a 2.5 µm paper filter (Whatman, Maidstone, UK, Type 42), and a 0.2 µm PTF E membrane filter (Whatman, Maidstone, UK). The filtration was performed with a filtration set equipped with a 47-mm glass filter funnel (PALL Corp., New York, USA) and vacuum pump. Three fractions of PM were collected: (i) 10–100 µm (large PM), (ii) 2.5–10 µm (coarse PM), and (iii) 0.2–2.5 µm (fine PM). Total PM ranged therefore from 0.2 to 100 µm. Before and after filtration filters were dried for 45 min at 60 °C in a drying cabinet, stabilised in the weighing room for 45 min, and weighed (balance XS105DU, Mettler-Toledo International Inc., Greifensee, Switzerland and deioniser gate, HAUG GmbH & Co. KG, Leinfelden-Echterdingen, Germany). The amount of wax dissolved in chloroform was assayed for each fruit sample in pre-weighed beakers after chloroform evaporation. The fruit area of each sample was determined on peeled skins (Image Analysis System, Skye Instruments Ltd., Llandrindod Wells, UK and Skye-Leaf software).

2.4. Statistics

The Shapiro-Wilk test was used to examine the normality of distribution. The main effects and interactions of location, distance to road and date of sampling on PM accumulation and wax content on apple fruits were analysed with ANOVA. ANOVA was also used to analyse the effect of location on PM accumulation and wax content on plum fruits. The results were subjected to an analysis of variance at a significance level of $p = 0.05$. The Duncan test was used to determine the significance of differences between the means of the main effects. The number of samples of individual parameters is given in the descriptions of the relevant tables.

3. Results

The accumulation of PM and wax content on apples differed by orchard location (proximity to roads with different traffic intensity) and sampling date. A summary of the main effects and their interactions is presented in Table 6. In the vast majority of cases, the examined parameters were significantly affected by orchard location and sampling date. The exceptions were the smallest PM fraction (0.2–2.5 µm), for which significance was not recorded for orchard location, and wax content, where sampling date was not significant. No significance was reported for distance from the road. Of the interactions tested, only the interaction between orchard location and sampling date was significant, with the exception of wax content (Table 6).

Table 6. Summary of the main effects and interactions of location (proximity to roads with different traffic intensity), distance to road, and date of sampling on particulate matter (PM) accumulation and wax content on apples, shown as significant (1) or not significant (0).

Parameter	Location	Distance to Road	Date of Sampling	Location × Distance to Road	Location × Date of Sampling	Distance to Road × Date of Sampling	Location × Distance to Road × Date of Sampling
Total PM	1	0	1	0	1	0	0
Fine PM (0.2–2.5 µm)	0	0	1	0	1	0	0
Coarse PM (2.5–10 µm)	1	0	1	0	1	0	0
Large PM (10–100 µm)	1	0	1	0	1	0	0
Surface PM ($_S$PM)	1	0	1	0	1	0	0
In-wax PM ($_W$PM)	1	0	1	0	1	0	0
Wax content	1	0	0	0	0	0	0

3.1. *Effect of Orchard Location, Sampling Date and Distance from the Road on PM Accumulation and Wax Content on Apples*

On average, the significantly higher accumulation of total PM and in-wax PM ($_W$PM) was recorded on apples from trees grown in the rural location and close to the local road, while lower PM depositions (by 43% and 33–35% for total PM and in-wax PM respectively) was noted on fruits from the orchard situated near the national road (Table 7). Fruits from the orchard in the rural location also accumulated the greatest amounts of PM fraction 2.5–10 µm and surface PM ($_S$PM), while for PM fraction 10–100 µm the highest deposition was recorded in the orchard adjacent to the local road. In the orchard near the national road, apples also accumulated the lowest amount of the fine PM fraction (0.2–2.5 µm), but the differences compared with other locations, although relatively high (9.5–15.5%), were not significant (Table 7).

Accumulation of PM (total PM, fine PM, coarse PM, large PM, surface PM, and in-wax PM) was on average significantly higher when apples were at harvest maturity stage (harvest period, sampled in October) than on the undeveloped fruits (sampled in June) (Table 7). When the locations were analysed individually, the highest PM accumulation (total PM, all PM size fractions and categories) was also recorded at harvest, with the only exception being PM of fraction 2.5–10 µm in the orchard located in the rural area. In June, the highest accumulation of total PM, all PM size fractions and categories (except 0.2–2.5 µm) was recorded in the orchard situated in a rural location, while at harvest maturity stage it was in the orchard next to a local road (Table 7).

The location of the orchard affected the wax content on apples (Table 7), which on average was significantly higher on apples harvested from orchards located close to the local road and in the orchard located in the rural area. On average, the date of harvest did not have a significant impact on the wax content on fruits. When the locations were analysed individually, no significant differences were recorded in June, while at harvest maturity stage a significantly higher wax content was recorded in the orchard in a rural area (Table 7).

Table 7. Effect of orchard location (proximity to roads with different traffic intensity) and sampling date on accumulation of total PM (0.2–100 µm), fine PM (0.2–2.5 µm), coarse PM (2.5–10 µm), large PM (10–100 µm), surface PM ($_{S}$PM), in-wax PM ($_{W}$PM), and wax content on apples. Data are means ± SE, n = 9 (3 replications × 3 distances to road), 18 (average for location: 3 replications × 3 distances to road × 2 sampling dates) and 27 (average for sampling date: 3 replications × 3 locations × 3 distances to road).

Orchard Location	Rural Location	Local Road	National Road	Avg. Sampling Date (B)
		Total PM [µg·cm^{-2}]		
June	30.56 (±3.09)	10.15 (±0.93)	13.67 (±1.07)	18.13 (±2.06) b [1]
Harvest	41.59 (±1.87)	62.02 (±4.25)	27.48 (±2.11)	43.69 (±3.22) a
Avg. location (A)	36.07 (±2.20) A [2]	36.08 (±6.63) A	20.57 (±2.03) B	LSD (A/B) = 8.74; (B/A) = 7.24
		Fine PM (0.2–2.5 µm) [µg·cm^{-2}]		
June	1.55 (±0.17)	1.78 (±0.16)	1.95 (±0.32)	1.76 (±0.13) b
August	3.72 (±0.37)	3.87 (±0.31)	2.83 (±0.19)	3.47 (±0.19) a
Avg. location (A)	2.64 (±0.33) A	2.83 (±0.31) A	2.39 (±0.21) A	LSD (A/B) = 0.92; (B/A) = 0.76
		Coarse PM (2.5–10 µm) [µg·cm^{-2}]		
June	10.87 (±1.41)	1.92 (±0.24)	2.99 (±0.31)	5.26 (±0.91) b
August	8.93 (±0.87)	15.97 (±0.67)	7.99 (±1.00)	10.96 (±0.85) a
Avg. location (A)	9.90 (±0.84) A	8.94 (±1.74) AB	5.49 (±0.79) B	LSD (A/B) = 3.05; (B/A) = 2.53
		Large PM (10–100 µm) [µg·cm^{-2}]		
June	18.15 (±1.59)	6.46 (±0.70)	8.73 (±0.67)	11.11 (±1.16) b
August	28.93 (±2.02)	42.18 (±3.80)	16.66 (±1.11)	29.26 (±2.49) a
Avg. location (A)	23.54 (±1.81) AB	24.32 (±4.72) A	12.69 (±1.15) B	LSD (A/B) = 6.96; (B/A) = 5.77
		Surface PM ($_{S}$PM) [µg·cm^{-2}].		
June	14.60 (±1.75)	2.42 (±0.40)	4.11 (±0.64)	7.05 (±1.22) b
August	19.60 (±2.36)	30.88 (±3.02)	11.66 (±0.86)	20.71 (±1.99) a
Avg. location (A)	17.10 (±1.54) A	16.65 (±3.75) AB	7.88 (±1.05) B	LSD (A/B) = 6.22; (B/A) = 5.15
		In-wax PM ($_{W}$PM) [µg·cm^{-2}]		
June	15.96 (±1.38)	7.73 (±0.78)	9.56 (±0.72)	11.08 (±0.89) b
August	21.98 (±1.88)	31.14 (±2.46)	15.82 (±1.41)	22.98 (±1.65) a
Avg. location (A)	18.97 (±1.35) A	19.44 (±3.10) A	12.69 (±1.08) B	LSD (A/B) = 5.84; (B/A) = 4.83
		Wax content [µg·cm^{-2}]		
June	449.5 (±5.58)	495.8 (±14.7)	456.8 (±12.2)	467.4 (±7.52) a
August	489.1 (±24.9)	502.3 (±8.68)	433.0 (±17.2)	474.8 (±11. 7) a
Avg. location (A)	469.3 (±13.3) AB	499.0 (±8.3) A	445.0 (±10.6) B	LSD (A/B) = 54.02; (B/A) = 44.73

[1] Different lowercase letters within a column indicate a significant difference between the sampling dates at $p < 0.05$ by the Duncan test. [2] Different uppercase letters within a row indicate a significant difference between the studied orchards at $p < 0.05$ by the Duncan test.

Distance from the road (expected main pollution source) had no significant effect on PM accumulation (total PM, all PM size fractions and categories) and wax content, but a slightly higher PM deposition was recorded on fruits from the orchard located closest to the road (first row from the road) (total PM, fine PM, large PM, and in-wax PM) (Table 8).

Table 8. Effect of distance to the road on accumulation of total PM (0.2–100 µm), fine PM (0.2–2.5 µm), (2.5–10 µm), large (10–100 µm), surface PM ($_{S}$PM), in-wax PM ($_{W}$PM), and wax content on apple fruits. Data are means ± SE, n = 18 (3 replications × 3 locations × 2 sampling dates).

Parameter	Distance A	Distance B	Distance C
Total PM [µg·cm^{-2}]	32.18 (±4.39) a [1]	29.72 (±4.25) a	30.83 (±4.98) a
Fine PM (0.2–2.5 µm) [µg·cm^{-2}]	2.77 (±0.27) a	2.42 (±0.27) a	2.66 (±0.32) a
Coarse PM (2.5–10 µm) [µg·cm^{-2}]	8.34 (±1.25) a	8.58 (±1.32) a	7.42 (±1.28) a
Large PM (10–100 µm) [µg·cm^{-2}]	21.07 (±3.11) a	18.73 (±2.89) a	20.76 (±3.69) a
Surface PM ($_{S}$PM) [µg·cm^{-2}]	14.07 (±2.27) a	13.48 (±2.67) a	14.09 (±2.91) a
In-wax PM ($_{W}$PM) [µg·cm^{-2}]	18.11 (±2.33) a	16.24 (±1.81) a	16.74 (±2.33) a
Wax content [µg·cm^{-2}]	464.5 (±11.7) a	476.3 (±11. 8) a	472.4 (±12.8) a

[1] Different lowercase letters within the row indicate significant difference at $p < 0.05$ by the Duncan test.

3.2. Effect of Orchard Location on PM Accumulation and Wax Content on Plums

The location of the orchard had no significant effect on PM accumulation (total PM, all categories and size fractions) on plums (Table 9). Fruits sampled from the orchard situated close to the national road accumulated slightly more total PM, PM of size fractions 10–100 µm and 0.2–2.5 µm, and surface PM ($_{S}$PM), but less of 2.5–10 µm. The wax content was significantly the lowest on plums from the orchard located in the rural area (Table 9).

Table 9. Effect of orchard location proximity to roads with different traffic intensity) on accumulation of total PM (0.2–100 µm), fine PM (0.2–2.5 µm), coarse PM (2.5–10 µm), large PM (10–100 µm), surface PM ($_{S}$PM), in-wax PM ($_{W}$PM), and wax content on plum fruits. Data are means ± SE, n = 3.

Parameter	Rural Location	Local Road	National Road
Total PM [µg·cm^{-2}]	56.57 (±2.68) a [1]	58.27 (±4.08) a	61.61 (±8.76) a
Fine PM (0.2–2.5 µm) [µg·cm^{-2}]	3.96 (±0.65) a	2.36 (±0.22) a	4.59 (±0.80) a
Coarse PM (2.5–10 µm) [µg·cm^{-2}]	8.28 (±0.94) a	7.06 (±0.94) a	4.92 (±1.06) a
Large PM (10–100 µm) [µg·cm^{-2}]	44.33 (±2.12) a	48.85 (±3.17) a	52.10 (±8.34) a
Surface PM ($_{S}$PM) [µg·cm^{-2}]	30.00 (±2.20) a	32.74 (±2.67) a	35.04 (±9.39) a
In-wax PM ($_{W}$PM) [µg·cm^{-2}]	26.57 (±0.60) a	25.53 (±1.66) a	26.57 (±1.98) a
Wax content [µg·cm^{-2}]	709.9 (±57.7) b	1247.6 (±134.0) a	1257.9 (±11.7) a

[1] Different lowercase letters within the row indicate significant difference at $p < 0.05$ by the Duncan test.

3.3. Comparison of PM Accumulation by Apples and Plums

In a comparison of PM accumulation by apples and plums sampled at the same physiological stage of fruits (harvest maturity) and the same distance from the road (first row from the road), PM deposition on plums was found to be significantly higher than on apples (Table 10). Plums accumulated more total PM, PM of size fraction 10–100 µm, and surface PM ($_{S}$PM). There were no differences in the accumulation of in-wax PM ($_{W}$PM) and smallest PM fraction (0.2–2.5 µm), while deposition of PM fraction 2.5–10 µm was significantly higher on apples. The plums were covered in a significantly greater amount of wax (Table 10).

Table 10. Effect of fruit species (apples, plums) on accumulation of total PM (0.2–100 µm), fine PM (0.2–2.5 µm), coarse PM (2.5–10 µm), large PM (10–100 µm), surface PM ($_{S}$PM), in-wax PM ($_{W}$PM), and wax content. Data are means ± SE, n = 9 (3 replications × 3 locations).

Parameter	Apple	Plum
Total PM [µg·cm^{-2}]	44.64 (±5.16) b [1]	58.81 (±2.99) a
Fine PM (0.2–2.5 µm) [µg·cm^{-2}]	3.65 (±0.33) a	3.63 (±0.45) a
Coarse PM (2.5–10 µm) [µg·cm^{-2}]	10.91 (±1.43) a	6.75 (±0.69) b
Large PM (10–100 µm) [µg·cm^{-2}]	30.08 (±3.88) b	48.43 (±2.88) a
Surface PM ($_{S}$PM) [µg·cm^{-2}]	19.78 (±2.88) b	32.59 (±2.98) a
In-wax PM ($_{W}$PM) [µg·cm^{-2}]	24.85 (±2.96) a	26.22 (±0.78) a
Wax content [µg·cm^{-2}]	465.8 (±20.2) b	1071.8 (±99.9) a

[1] Different lowercase letters within the row indicate significant difference at $p < 0.05$ by the Duncan test.

4. Discussion

4.1. PM Accumulation by Apples and Plums Is Not Affected by Road Type and Distance from the Road

Safety is the main criterion when assessing food quality. Fruits, often eaten raw, are considered to be less endangered by pollutant accumulation than vegetables, especially root vegetables [36]. It has been already been demonstrated that translocation of heavy metals from the soil to fruits is relatively unlikely [17,18,37]. Nonetheless, the content of various pollutants in fruits from trees grown near urban and/or industrial areas might exceed safety limits [11,19,23,36]. The most important source of pollution for fruits seems to be airborne PM, emitted e.g., from road sources [12,29,38,39]. In the present study, apples and plums fruits accumulated considerable amounts of PM. The PM accumulation per area unit by fruits was at the same level or more often even higher than that reported for foliage of urban shrubs and trees when samples were examined using the same analytical method as in this study, e.g., [40,41]. At this level of PM accumulation, foliage of shrubs and trees is already considered to be an effective air filter, e.g., [40,41]. To some extent this is not surprising, since the fruits of *Ricinus communis* L. have already been described as better biomonitors of trace elements concentrations in the atmosphere (Cu, Pb, Zn) than leaves [42]. This poses a serious concern about the safety of fruits produced in locations subject to significant anthropogenic pressure, such as urban gardens and orchards, industrial areas and especially sites close to roads. This problem has already been recognised in China, for example, where the government has set environmental requirements for air quality (<300 $\mu g \cdot m^{-3}$ of total suspended particulates) in fruit production areas (Free food apple producing environmental conditions, NY 5013-2001). In the present study, three roads with varying levels of traffic (Tables 1 and 4) were expected to be the main source of PM in examined orchards. It has previously been shown that road transport sources, both exhaust and non-exhaust, account for significant PM emissions [11,28,30–32,43] and may be the reason for pollution accumulation in edible plants [29,38,39]. Road PM emission is increased by the number and size of vehicles [32,44–46], average speeds and sudden changes in speed [44,46,47], and is higher in spring and summer than in winter, and higher during the week than at weekends [28]. The motorway induces a contamination of the surrounding environment up to 320 m away, but with the maximum pollution recorded at between 5 and 20 m [28,48]. Zhu et al. [49] suggested, that exposure to PM is significant within 100 m downwind of major traffic sources such as roads. It was therefore expected that PM accumulation would be highest in orchards located close to busy national roads and on fruits sampled from the rows closest to the roads. Surprisingly, in the case of apples, although accumulation near the national road (very high traffic density) was high compared with tree foliage in urban areas, it was the lowest of the three locations examined, while no impact of the neighbouring road was found on the level of PM accumulation in plums. Also the average ambient concentration of PM in the apple orchards (measured once a month, Table 3) differed only slightly. These results suggest that PM accumulation on fruits depends primarily on the activities undertaken in the orchard itself and sources of PM pollution other than the roads located in the orchard's immediate vicinity. The concentration of ambient PM in an orchard can fluctuate rapidly and reach high levels in a short period of time. The main PM source in the orchard in the rural area could be nearby construction site. The orchard adjacent to the local road was managed organically, with the cultivation of a large number of apple varieties harvested on different dates (from mid-September to late October) and the application of plant protective agents authorised for use in organic production, which often increase the surface stickiness of the fruits. Late harvest may also result in the accumulation of airborne pollutants created by domestic heating, which has already been recorded in apple skins [11,15]. Furthermore, in the present study, the accumulation of PM on apple fruits was also not affected by distance from the road. These results confirm the findings of Bakirdere and Yaman [50] who demonstrated that accumulation of airborne Cd and Pb does not decrease with increasing distance from roads but, in contrast to the present study, with decreasing traffic densities. On the contrary, Mori et al. [51] and Popek et al. [52] showed that PM accumulation on the foliage of shrubs and trees is highest in the case of plants grown nearest roads compared with those at greater distances. We assume, that lack of difference in PM accumulation between fruits from trees growing at different distances from roads can be explained by the large spacing between trees in the orchard. Instead of being filtered or stopped by vegetation, as occurs in the case of roadside or park greenery [53], polluted air penetrated deeper into the orchard, with PM evenly distributed over a larger orchard area. The high velocity and turbulence of the air coming from roads further increased PM dispersion in the orchards [28]. It seems that planting orchards relatively close to the road is burdened with less risk than previously thought. The results of this study suggest that if an orchard is at least 10–20 m from the road and the polluted air blown from the road is not slowed down or stopped by physical barriers, the accumulation of PM by fruits is similar irrespective of the trees' location in the orchard up to at least the tenth row of trees. These barriers can be buildings or vegetation, and obviously increasing the height and width of a barrier will decrease the pollutant contents in the edible part of fruits [54]. The best solution would be trees and shrubs because they are porous enough to allow penetration, instead of deflecting the air stream above the barrier [55]. These new data do not discount the fact that roads are an important source of air pollution in fruit production, but do indicate that other factors may be equally or even more important. However, it should be remembered that in this work we did not analyze the chemical composition of PM accumulated by fruits, which may differ depending on the distance from the road.

4.2. PM Quantity on Fruits Increases in Successive Months

The level of PM accumulation on apples increased between sampling dates, confirming previous findings for leaves and needles [56]. In the present study, however, changes in PM accumulation between the sampling dates primarily resulted not from the constant accumulation of PM generated during road transport, but from events within the orchard or its immediate vicinity. In the orchard located in the rural area, only a slight increase in PM accumulation was recorded between fruits sampled in July and September. This was due to a very high accumulation of PM emitted in June–July as a result of a great deal of construction work nearby. The construction was completed in early August, about three weeks after the first fruit sampling, and thus only had a limited effect on the amount of PM on fruits harvested in October. In contrast, in the extensive orchard adjacent to a busy local road, a peak of PM accumulation was noted in September-October when fruits of different apple varieties were successively harvested as they matured. The 'Gloster' variety of trees examined in this study were grown next to the main path in the orchard that is frequently used by mechanical devices (tractors, sprayers), and their fruits were some of the last to be harvested, thus they accumulated PM generated in the orchard during all the maintenance work and the whole harvest. Only the orchard running alongside the national road reflected the real impact of the road on changes in PM accumulation by fruits during their ripening. In this location, PM accumulation was also greater at harvest (October), but the increase between sampling dates was much less pronounced than in the orchard near the local road.

The higher PM load on apples at harvest in all orchards may also result from the fruits' development stage and standard maintenance work carried out in the orchards. In July the apples were still small and had short petioles, therefore they were protected from ambient air by leaves, which at that time were fully developed and had reached their maximum size. After the first sampling, the trees were pruned to reveal the fruits and increase their exposure to light. This undoubtedly resulted in increased accumulation of PM from fertilisers, ripening agents and pesticide applications [7,34], as well as from traffic and other sources not associated with orchards. This suggests that the apple varieties harvested later would be much more likely to have increased PM accumulation and should be planted away from houses and farm buildings, roads and vehicular routes in the orchards.

The orchards studied differed in PM size fractions, whose accumulation on apples increased most between sampling dates. In orchards facing local and national roads, the greatest increase was recorded for coarse PM (2.5–10 μm) followed by large PM (10–100 μm), while in the case of the orchard in the rural area, the greatest difference was noted for fine PM (0.2–2.5 μm) also followed by large PM (10–100 μm). Unfortunately, PM particles smaller than 10 μm (fraction 0.2–10 μm in this study) are also the most dangerous to human health [1] as they are the main carriers of toxic trace elements and polycyclic aromatic hydrocarbons [6,18]. To some extent, such a great increase in the deposition of fine and coarse PM is surprising because, in the case of leaves, regardless of the species and location studied, the largest increase in weight of accumulated PM throughout the growing season is associated with the fraction of the largest PM (10–100 μm) [56]. On apples sampled at harvest, the average share of PM fraction 10–100 μm (69%, 68% and 61% in orchards in the rural area and close to local and national roads respectively) was also lower than that recorded on leaves and needles [40,41,56]. It has been shown that road transport mainly emits the smallest PM, with $PM_{0.5}$ accounting for 56% of the PM_{10} mass and $PM_{2.5\text{-}10}$ representing just 12% [57]. In this work PM belonged to the smallest fraction (0.2–2.5 μm) maked up just 8.5% of all accumulated PM on apples. This may be further evidence that although fruits certainly adsorbed considerable amounts of PM emitted from the road (especially in the orchard close to the national road), in this study it is highly likely that most of the PM accumulated by fruits came from sources other than transport.

In contrast to PM, the amount of wax on fruits did not change between sampling dates and was similar in all orchards. However, in all the examined locations, the fruits sampled in October accumulated a significantly higher amount of PM deposited in wax ($_{W}$PM). It can therefore be assumed that the proportion of lipophilic pollutants increased, e.g., polycyclic aromatic hydrocarbons from domestic heating and most likely vehicles operating within orchards and on surrounding roads [15,31]. The share of PM embedded in wax was highest in apples harvested in the orchard near the national road, which confirms that although transport was not the main source of PM in this study, at this location its contribution was greater than in other orchards.

4.3. Plums Accumulate More PM than Apples

Comparing PM accumulation by apples and plums from trees grown in orchards located in very similar conditions (distance to roads with similar traffic intensity), a significantly higher PM load was found on plums, although plums were harvested more than three months earlier than apples. These results suggest that in orchards located close to busy roads (or other continuous sources of PM emissions), the selection of crop is important. Species/cultivars with a tendency for less PM accumulation and early harvest date should be planted. The difference in accumulation of airborne pollutants has already been shown with leaves of various trees and shrubs, where their morphology, density and inclination angle play a decisive role [13,40,58,59]. Fruits examined in this study were also morphologically different. Plums were smaller and covered in a much thicker wax layer, which however did not result in a higher accumulation of in-wax PM ($_{W}$PM). Higher PM accumulation by plums was found instead from increased adsorption of large PM (10–100 μm) and surface PM ($_{S}$PM). Although the structure and chemical composition of the wax layer was not studied in this work, it was assumed that the wax on the plum surface was sticky enough to absorb and retain hydrophilic PM on its surface. Furthermore, apples and plums differ in the density and size of stomata and lenticels, which are well known places of increased accumulation of airborne pollutants [20,58]. It was shown, that heavy metal uptake by foliar surfaces occurs through stomata, cuticular cracks, lenticels, ectodesmata and aqueous pores [18]. Larger lenticels on apples may have resulted in easier penetration of PM inside fruit than was the case in plums. However, the size and density of stomata and lenticels on fruits differ greatly between species and varieties, fruits age and most probably microclimatic conditions [60–62], and therefore their role in PM accumulation should be investigated further. Tree height may also be relevant. According to von Hoffen and Säumel [54], the greater sampling height of the elder berries compared to other berries reduced the trace metal content. In this study, however, fruits from higher trees (plums) accumulated more PM than fruits from lower trees (apples). Trees of both species also differ in plant architecture, with plums having a more complex crown, which most probably increases PM accumulation [63,64]. The cultivation of apple and plums differs in the number of treatments. Interestingly, more chemical treatments and mechanical pruning of trees (exposing the fruit to light) are taking place in apple orchards (Tables 2 and 5). Nevertheless, PM accumulation was higher on plums, which may suggest that the amount of PM on the fruit is determined by their morphological features, most likely the waxy layer. To the best of the authors' knowledge, a comparison of PM accumulation by edible fruits has not yet been presented in the literature. It is believed that the different content of toxic trace elements and polycyclic aromatic hydrocarbons in fruits [11,19,23,36] is primarily due to the different accumulation of PM. Knowledge about the factors that affect PM deposition on fruits could be used to improve their quality.

4.4. Washing Fruits Removes Substantial Amount of Accumulated PM

Once PM is in the orchard, it is not easy to limit its deposition on fruits. One option may be fruit bagging, but this treatment is expensive, laborious and changes the properties of fruits [65]. The toxicity of PM already deposited on the surface of fruits can be reduced by washing treatments. In the present study, gentle rinsing of apples and plums for one minute with distilled water resulted in the removal of 44% and 55% PM respectively. This means that intensive washing of fruit before consumption should definitely remove more than half of the accumulated PM, and thus should be a habit when fruit is from an unknown source [54]. Von Hoffen and Säumel [54] demonstrated that the consumption of non-vegetable fruits growing in inner-city sites in Berlin does not pose a risk to human health if the fruits are thoroughly washed. It should be noted that water will only wash away pollutants adsorbed to the fruit surface [17]. This also applies to pollutants carried by PM, e.g., Fe and Zn are harder to wash off than Cd and Pb because they tend to penetrate the plant surface more deeply [25]. Polycyclic aromatic hydrocarbons are particularly difficult to remove from fruit surfaces because their lipophilic nature means that they are embedded almost exclusively in the epicarp, especially wax [15]. Schreck et al. [17] and De Temmerman et al. [16] demonstrated that washing removes a maximum of 25% of total Pb-rich and Cd-rich particles, which might suggest that a significant amount of PM is deposited in the cuticular wax. Fortunately, according to Paris et al. [15] airborne pollution is rarely transferred to the mesocarp, because skin is an efficient hydrophobic protective barrier. Peeling removes most of the cumulative pollution by fruits from the air [66], but this reduces their nutritional value.

5. Conclusions

The results obtained in this study suggest that PM is deposited on apples and plums harvested from orchards located close to roads with different traffic intensities. The amount of PM deposited on fruits is comparable to or higher than that on the foliage of trees and shrubs growing in polluted city centres. PM accumulation by fruits depends primarily on: (i) the plant species and most probably the varieties (fruit morphology, especially the amount of wax), (ii) harvest period (late harvest is associated with the negative impact of domestic heating), (iii) maintenance work in the orchard (early pruning of trees will expose fruits to PM), (iv) adverse events during fruit development taking place in the orchard and its immediate surroundings (construction works and fires), and (v) cultivation method (ecological preparations can increase the viscosity of the fruit). The impact of roads seems less significant, nevertheless PM from transport, despite being accumulated in smaller amounts, may be more toxic. Washing with water removes about 50% of the PM accumulated by the fruit. The results of this study suggest that PM may be an important source of fruit contamination by substances that are hazardous to human health.

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