

Research Article

Distribution and Fractional Composition of Petroleum Hydrocarbons in Roadside Soils

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Total petroleum hydrocarbon (TPH) concentrations and their fractional composition (medium fraction: n-alkane chain-length C15 to C27, heavy fraction: >C27) were determined at distances from 1 to 60 m from roads and at soil depths from 0.5 to 15 cm. The traffic intensities were up to 25000 vehicles per day. Soil TPH concentrations were highest within 15 m distance (665 and 3198 mg kg⁻¹ at the windward and leeward sides, resp.), followed by a rapid drop to background values beyond (196 and 115 mg kg⁻¹ in 60 m distance at the windward and leeward sides, resp.). The data variability was lowest at distances of 1 m and highest within tree plantations at distances of 15 m from the road. The TPH concentrations decreased with depth but were significantly higher than the background at all depths investigated. A principal component analysis revealed a positive relation between the medium-to-heavy fraction ratio and soil depth. A fractional differentiation of hydrocarbons with distance from road was not observed. It was concluded that the assessment of the potential of hydrocarbons to translocate, accumulate, or degrade in soil necessitates their subdivision into fractions based on their physicochemical and metabolic properties.

1. Introduction

The widespread use of hydrocarbons in fuels causes their predominance among organic atmospheric pollutants, and petroleum products are the major source of anthropogenic hydrocarbon pollutants found in the atmosphere [1–3]. Total petroleum hydrocarbons (TPH) from fossil sources are not readily biodegradable as compared to biomass or soil organic matter, which have been demonstrated to be consumed during hydrocarbon degradation [4, 5]. Once deposited to the surface, hydrocarbons may persist and bioaccumulate in environmental media [6] and infiltrate into groundwater aquifers via leaching or into surface aquifers by runoff with severe effects on plants [7, 8], humans, and animals [9, 10]. Organic contaminants in roadside soils have been receiving considerable attention as a result of traffic intensity [1, 2, 11, 12]. Hydrocarbon deposition to ecosystems is of more complex nature, because it is influenced by meteorological

and further peripheral conditions, like wind, geomorphology, road construction, buildings, or vegetation, resulting in atmospheric dilution, turbulent exchange, possible wind channeling, and so forth [12].

Apart from studies dealing with individual substances, TPH have been treated in the literature as one class of substances so far. However, as a diverse mix of numerous individual aliphatic hydrocarbons, components of petroleum products also behave individually in the environment. This consideration gives rise to our assumption that an investigation of translocation of TPH through and accumulation of TPH in environmental media must be based on their subdivision at least into fractions of substances with similar physical, chemical, and metabolic properties.

It is the aim of this study to prove the hypothesis that peripheral conditions, in particular vegetation, influence (I) amount and spatial variability of TPH in roadside soils and that the fractional composition of petroleum hydrocarbons

TABLE 1: General site characteristics.

	Traktorostroiteley avenue	Pushkinskaya street	M18 Kharkov-Yalta
Traffic density ¹	1027	774	575
Soil texture ²	Clayey loam	Loamy clay	Sandy-loamy silt
Org. C (%)	2.65 ± 1.87	2.41 ± 1.39	2.24 ± 0.97
Na (%)	0.45 ± 0.05	0.50 ± 0.26	0.52 ± 0.12
Mg (%)	0.43 ± 0.19	0.40 ± 0.16	0.43 ± 0.29
Al (%)	3.42 ± 1.33	3.20 ± 1.43	3.47 ± 2.23
Si (%)	36.96 ± 3.06	37.62 ± 2.33	37.10 ± 4.83
K (%)	1.65 ± 0.40	1.34 ± 0.38	2.29 ± 1.82
Ca (%)	1.91 ± 0.60	1.39 ± 0.25	1.32 ± 0.17
Mn (%)	0.05 ± 0.02	0.05 ± 0.01	0.06 ± 0.01
Fe (%)	1.95 ± 0.77	2.20 ± 0.61	1.91 ± 1.13

¹Vehicles per hour, medium values from October 2010, ²according to Arbeitsgemeinschaft Boden [13], arithmetic mean values ± standard deviation.

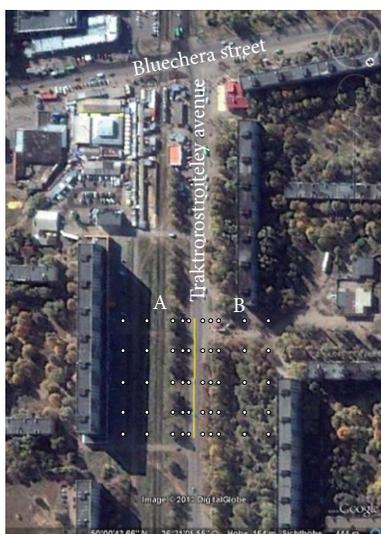


FIGURE 1: Map of the Traktorostroiteley avenue sampling location; A: windward side; B: leeward side; sampling points are marked as white dots, and the yellow line depicts the 100 m central section of the road. Image source: Google Earth, retrieved 30.08.2013.

changes with (II) distance from the road and (III) with soil depth.

2. Material and Methods

Soil samples were collected at distances of 1, 6–8, 15, 40, and 60 m and at depths from 0.5 to 15 cm in October 2010 from the Traktorostroiteley avenue in Kharkov (50°0'41''N, 36°21'3''E). Five replicates along a 100 m section at the windward and the leeward sides of the road, as referred to the long-term main wind direction (sides A and B, resp.), were sampled. A schematic map of the sampling location is given in Figure 1. At distances 3 of 8 m (windward side A) a single row of birch trees (distance between trees from 6 to 10 m) and 4 m (leeward side B) from the road a double row of maple trees (distance between trees from 4 to 6 m) were planted.

In addition, the Pushkinskaya street ($n = 4$) and the M18 highway (pooled samples taken in October 2011, $n = 5$) were taken into consideration for multivariate statistical analysis. An overview of the sampling sites is given in Table 1. The 60 m samples served as background level control. The soil type was Chernozem, developed on Loess at all sampling locations. The long-term mean annual precipitation in Kharkov is 520 mm, and the long-term mean annual temperature is 7.5°C. Plant litter and biomass are being removed from the surface twice per year in spring and in fall, and an organic soil horizon has not formed on top of the mineral mull horizon.

The samples were air-dried immediately after sampling to prevent microbial degradation of hydrocarbons. Total element concentrations were determined using energy-dispersive X-ray fluorescence spectroscopy (x-supreme EDXRF analyser, Oxford Instruments, UK). Total soil carbon was determined by dry combustion with O₂ at 1050°C using a CNS Analyser (Elementar, Germany). Carbonates were determined volumetrically as CO₂ after HCl treatment. Soil organic carbon was determined by subtraction of carbonate-C from total C.

TPH concentrations in soil were determined gravimetrically after triple extraction of 20 g sieved soil (<2 mm) with 50 mL hexane p.a., cleanup over a short aluminum oxide column, and solvent evaporation to constant weight at 40°C. TPH and fractional composition of petroleum hydrocarbons of selected samples were further determined using GC-MS (Carlo Erba, Fisons) after triple extraction of 2 g soil with 5 mL hexane for residue analysis, cleanup over a short fluoro-sil column, and solvent evaporation under hydrocarbon-free nitrogen at 20°C to a sample volume of 10–100 µL, depending on the results of the gravimetric hydrocarbon determination. Commercial diesel fuel and Hewlett Packard alkane standard Part number 18710-60170 were used for quantification and identification of total hydrocarbons and n-alkanes, respectively. We used an SGE HT8 column at 2 mL min⁻¹ helium flow with 1 µL split injection at a split ratio of 20:1. Injection temperature was 300°C. The column temperature was 70°C during injection and was increased after 1 min with a heating rate of 30 K min⁻¹ and held constant for 20 min after reaching 320°C. For detection we used

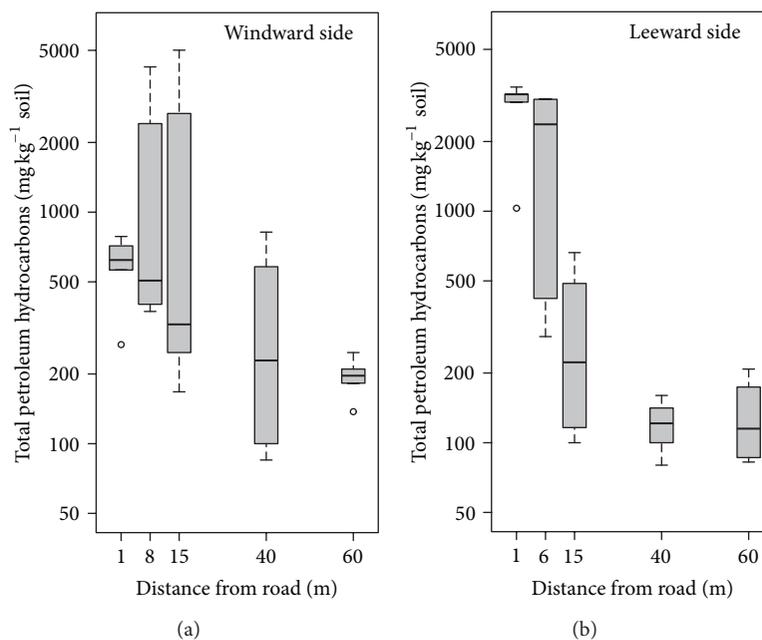


FIGURE 2: Boxplot of combined GC-MS and gravimetric datasets of TPH concentrations (mg kg^{-1} dry soil) at the windward and leeward sides depending on the distance from road, Traktorostroiteley avenue; bold lines: median values, boxes: upper and lower quartiles, whiskers: data ranges, and circles: outliers.

positive electron impact ionization (EI+) at 70 eV and MS full scan mode with m/z ranging from 50 to 250. A medium (C15 to C27) and a heavy fraction (>C27) were quantified in the chromatograms using the total ionic current (TIC) after elimination of siloxane signals resulting from column bleeding.

2.1. Statistical Analysis. Box-whisker-plots were used to depict median values (bold lines) and variation (upper and lower quartiles in boxes, data ranges as whiskers, outliers as circles) of TPH concentrations over distance from road. Principal component analysis (PCA) was performed to extract main components explaining the total data variability, where PCA biplots were used to identify correlations between variables. For PCA we used z -transformed gravimetric and GC-MS TPH concentrations, soil organic carbon, Al, Fe, and Si concentrations, the distance from road, sampling depth, and the medium-to-heavy fraction ratio. Bivariate regression analysis, Pearson's product moment correlation, and Student's t -tests were applied to check for statistical significance. We used the R software suite for all statistical calculations.

3. Results

The highest TPH concentrations were observed within a 15 m strip along the road, followed by a rapid drop to background values beyond. At the leeward side, the TPH concentrations in 1 m distance from the road amounted to a median value of 3198 mg kg^{-1} and significantly (t -test, $P < 0.01$) exceeded the value of 665 mg kg^{-1} at the windward side by a factor of 4.8. In 60 m distance from the road, the TPH concentrations

amounted to 196 and 115 mg kg^{-1} at the windward and leeward sides, respectively, which did not differ significantly (t -test, $P = 0.14$). The data variability was lowest at a distance of 1 m and highest at a distance of 15 m from the road, where a general trend of decrease could be observed beyond 15 m (Figure 2).

Within the soil profiles of the contaminated 15 m strip, the TPH concentrations decreased with depth but were significantly elevated compared to the control at all depths investigated (t -test, $P < 0.05$, Figure 3). The highest TPH concentrations were observed within the top 5 cm of the soil profile.

Aliphatic hydrocarbons were the dominating organic contaminant of the soils, and the sum of PAHs ranged from 0.11 to 3.61% of the concentration of aliphatics. A list of individual compounds determined and of the unresolved complex mixture (UCM [14]) is given in the appendix. The medium-to-heavy fraction ratio amounted to 0.950, 0.856, 1.637, 0.972, and 0.759 for 1, 6–8, 15, 40, and 60 m distance from the road, respectively, as well as to 0.972, 2.966, and 2.097 for 0.5, 10, and 15 cm sampling depth, respectively.

The first and second components of a principal component analysis (PCA) explained 47.2% and 22.1% of the total variance, respectively. Subsequent regression analyses revealed a significant positive relation between the gravimetric and the GC-MS TPH concentrations ($r = 0.83$, $P < 0.01$), as well as a significant positive relation between the medium-to-heavy fraction ratio and soil depth ($r = 0.70$, $P < 0.05$). Both gravimetric (Figures 2 and 4) and GC-MS (Figure 4) TPH concentrations decreased with distance from the road. The medium-to-heavy fraction ratio did not correlate with the distance from the road ($r = 0.10$, $P = 0.84$). There was

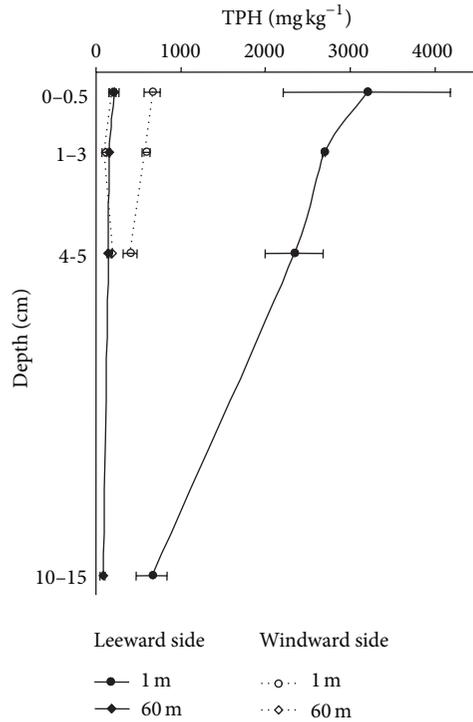


FIGURE 3: Dependence of TPH concentrations (arithmetic mean values \pm standard deviation) from the soil depth at the windward and leeward sides in 1 m and 60 m distance from road, Traktorostroiteley avenue.

a significant positive correlation between soil organic carbon, Al ($r = 0.79$, $P < 0.01$), and Fe ($r = 0.63$, $P < 0.05$), but a significant negative correlation between Si, soil organic carbon ($r = -0.78$, $P < 0.01$), Al ($r = -0.97$, $P < 0.01$), and Fe ($r = -0.91$, $P < 0.01$). Soil organic carbon did not correlate neither with gravimetric ($r = 0.11$, $P = 0.75$) nor with GC-MS TPH concentrations ($r = 0.20$, $P = 0.26$) (Figure 4).

Multiple regression analysis revealed a significant influence of traffic intensity (F -test, $P < 0.05$), distance from road (F -test, $P < 0.001$), sampling depth (F -test, $P < 0.001$), and period of road exploitation (F -test, $P < 0.001$) on total TPH concentrations.

4. Discussion

The TPH concentration in soils depends on traffic intensity [1, 2, 11, 12], as well as on meteorological and on further peripheral conditions, like wind, geomorphology, road construction, and buildings, or on vegetation. Hence, TPH amounts along roadside soils reported in the literature vary widely, reaching maximum levels of approximately 1100 mg kg⁻¹ soil in 1 m distance from a German freeway (50000 vehicles per day, [12]) and up to approximately 19500 mg kg⁻¹ soil in Nigeria [1]. The maximum levels found in this study amounted to 3198 mg kg⁻¹ soil at a traffic intensity of up to 25000 vehicles per day. In particular, for perpendicular approaching wind, markedly higher atmospheric pollutant concentrations at the leeward side and slightly lower concentrations at the

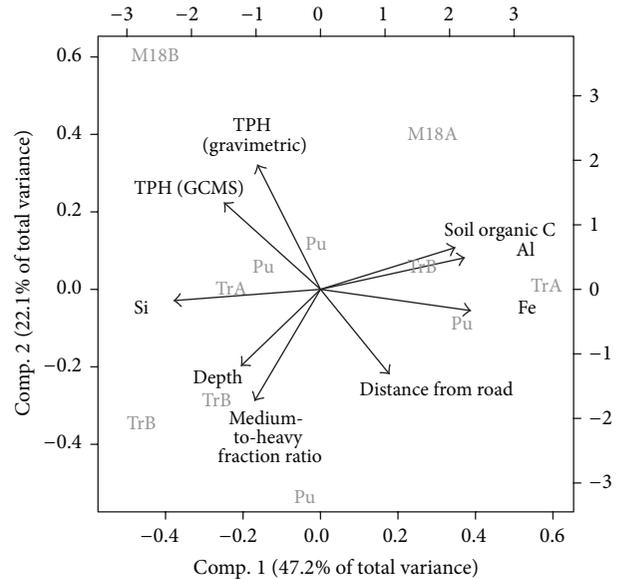


FIGURE 4: Biplot of PCA results after z-transformation; TPH: total petroleum hydrocarbons, Pu: Pushkinskaya street, Tr: Traktorostroiteley avenue, and M18: highway; letters A and B denote sampling windward and leeward sides, respectively.

windward side were observed by Gromke and Ruck [15], which may explain the higher soil TPH concentrations at the leeward side here. Unger and Prinz [12] reported that background levels were reached at a distance of 10–25 m from the road, which could be confirmed by the findings of this study (Figure 2).

With respect to their environmental impact, roads represent a linear source of contamination [12], and consequently low variation at given distance along the road may be expected under homogeneous conditions of TPH translocation, which increases as meteorological and further peripheral conditions vary within that given distance from road. In the present study, high values and low variation of TPH concentrations in 1 m distance from the road might be due to the influence of splash water carrying high amounts of hydrocarbons from the road surface and due to undisturbed by trees atmospheric hydrocarbon distribution. Surface runoff as major source of hydrocarbon input into soil can be excluded due to the presence of a curb. Ries and Eichhorn [16] reported reduced wind speeds within a vegetated street canyon with a slight increase of pollutant concentrations in the atmosphere. Tree plantings reduced the air exchange between street canyons and the ambience, where in comparison to treeless street canyons higher overall pollutant concentrations and lower flow velocities were measured by Gromke and Ruck [15]. Hence, wind channeling might promote long-distance hydrocarbon transport, whereas buildup of air in the vicinity of trees might cause longer dwell times of hydrocarbon containing air and, thus, may kinetically promote particulate sedimentation or direct adsorption of hydrocarbons to the surface before and reduce hydrocarbon deposition behind the tree rows. Unger and Prinz [12] reported a 35% increase of the TPH concentration before and

TABLE 2: Concentrations of individual components (in mg kg⁻¹ soil) of selected soil samples.

	Traktoroostroiteley avenue					M18		Pushkinskaya street			
	Windward		Leeward			Windward	Leeward	Leeward			
Distance from road (m)	1	6	60	1	1	1	1	1	15	40	100
Soil depth (cm)	0.5	0.5	0.5	10	15	0.5	0.5	0.5	0.5	0.5	0.5
TPH (GC-MS)	268	287	197	1919	1395	1273	3798	1185	312	453	108
Phenanthrene	0.39	0.62	0.79	4.90	1.07	0.67	13.7	2.49	1.76	1.35	0.10
Naphthalene						0.12				0.11	
Acenaphthylene							10.5		0.11		
Acenaphthene									0.19		
Fluoranthene	0.29	0.87	0.50	7.38	2.97	0.41	43.4	2.44	0.85	0.41	0.29
Pyrene	0.14	0.59	0.26	4.07	1.62	0.19	25.9	1.43	0.51	0.18	0.16
Benzo(a)anthracene				2.10	0.85		19.8	0.66	0.19		
Chrysene	0.08			1.99	0.67		24.0	0.76	0.20		0.05
PAH Sum	0.90	2.08	1.55	20.4	7.18	1.39	137	7.77	3.81	2.04	0.59
n-Pentadecane					0.17	0.23	0.78		0.25	0.33	
n-Hexadecane		1.04		3.21	1.15	1.20	6.85		1.25	1.79	
n-Heptadecane		0.60		1.73	0.55	0.83	7.14		1.58	1.20	
n-Octadecane	0.73	1.20		4.15	1.26	2.04	20.4		4.02	2.18	
n-Nonadecane	0.36			2.64		0.96	19.4		1.72	1.44	
Icosane	0.39	1.23		3.98	1.01	1.79	11.8		2.58	1.23	
Heneicosane	0.62	1.09		8.06	3.93	3.65	16.7		7.11	1.24	
Docosane	1.45	0.62	2.67	38.1	21.4	16.7	8.45	49.2	10.5	2.59	1.19
Tricosane	2.53		2.28	91.4	52.4	48.8			3.76	4.08	2.28
Tetracosane	3.41	0.16	1.25	119	72.1	89.4	12.1		2.70	5.20	3.33
Pentacosane	4.46	0.63	0.25	111	70.0	101		15.6	4.10	6.39	3.61
Hexacosane	4.49	0.32	0.69	89.2	60.0	95.4		20.1	3.12	7.14	3.29
Heptacosane	5.57	1.23	0.18	57.5	40.6	65.2		25.9	3.77	8.31	2.58
Octacosane	4.32	0.66	1.04	29.7	23.3	36.9		27.6	1.76	7.95	1.45
Nonacosane	6.15	3.90	0.41	2.21	13.1	19.8	197	29.0	3.57	10.6	1.25
Triacontane	3.47	0.64	1.08	7.01	6.24	9.05		24.3	0.87	7.74	0.78
Hentriacontane	4.65	4.85	0.15	5.71	4.30	7.28	111	23.1	5.05	10.8	0.76
Dotriacontane	1.29		5.65	2.23	1.87	4.02	33.0	15.8		4.82	
Tritriacontane	1.43		11.3	2.24	1.07	2.44	21.4	14.3		4.71	
Tetratriacontane	2.10		3.20	3.70	1.54	1.03		6.29	1.31	2.57	
Pentatriacontane	2.68					0.93		1.47	1.39	1.33	
UCM	216	267	165	1316	1012	763	3194	914	248	357	87
%UCM	81	93	84	69	73	60	84	77	79	79	80

Empty cells: below detection limit (PAH: 0.05 mg kg⁻¹; alkanes: 0.15 mg kg⁻¹); UCM: unresolved complex mixture [14].

a 90% reduction of the TPH concentration behind a forest belt. While the median TPH concentrations dropped with distance from the road, the presence of trees, hence, increased their spatial variability. Nonsignificant TPH concentrations in 60 m distance from the road at the windward and leeward sides point to their natural background (Figure 2). It should be noticed here that TPH concentrations at the windward side tended to exceed those of the leeward side in 60 m distance, which could not be expected in case of their translocation with prevailing winds. The insignificant relation between TPH concentration and traffic intensities found in this study points to the high relevance of meteorological and further peripheral local conditions.

Unger and Prinz [12] reported elevated TPH concentrations compared to the background level in a soil depth of 10 cm. In our study, we observed significantly elevated TPH concentrations down to our maximum sampling depth of 15 cm, indicating vertical translocation of hydrocarbons into the soil profile (Figure 3). The positive relation between the medium-to-heavy fraction ratio and soil depth as revealed from PCA (Figure 4) points to enhanced mobility of medium hydrocarbons and to their accelerated translocation into the soil profile, as compared to the heavy fraction. This phenomenon might be explained by decreasing with chain-length hydrocarbon solubility in water and biodegradability, as well as by increasing with chain-length adsorption to

the soil matrix, resulting in enhanced retention of heavier hydrocarbons along the path of water movement and in their relative accumulation in the upper soil. The enhanced mobility of the medium hydrocarbon fraction further imposes a risk to its movement into deep soil, where accumulation may be promoted by unfavorable conditions for biodegradation. A fractional differentiation of hydrocarbons along their atmospheric distribution path could not be observed within the 60 m strip of investigation.

Stabilization of soil organic matter by mineral surfaces, in particular by polymeric Fe and Al species, has been reported by Mikutta et al. [17]. In our study, soil organic carbon was associated with mineral phases rich in Fe and Al but was negatively correlated with mineral phases rich in Si. High concentrations of soil organic carbon, however, were not specifically associated with high TPH concentrations, indicating that the accumulation of petroleum hydrocarbons in soil was most likely controlled by site history and by peripheral conditions.

5. Conclusions

It can be concluded from the present results that petroleum hydrocarbons, presenting a mixture of numerous individual substances, behave nonuniformly in soil. To assess their potential to translocate, accumulate, or degrade in soil, it is necessary not only to determine total petroleum hydrocarbon concentrations but also to further subdivide them into fractions based on their physicochemical and metabolic properties.

Appendix

See Table 2.

Conflict of Interests

Larysa Mykhailova, Thomas Fischer, and Valentina Iurchenko have no conflict of interests.

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