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Assessment of soil and grass Polycyclic Aromatic Hydrocarbon (PAH) contamination levels in agricultural fields located near a motorway and an airport

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Abstract – Along motorways and airports, PAHs produced by vehicle exhausts can be deposited on grass and soil, leading to a risk of contamination through the food chain via the ruminant. Grass and soil were collected near the verge of a road (10 m) and a take-off strip (50 m). Analyses detected the 16 PAHs listed by the US Environmental Protection Agency in both sites and both grass and soil. The motorway site is about four times more contaminated (188 ng/g DW for grass) than the airport site (69 ng/g DW for grass) and carcinogenic compounds are present in both grass and soil. Furthermore, the part consisting of low molecular weight, and four and less than four ring compounds is higher in grass (36%) than in soil, where high molecular weight and more than four ring compounds represent 86% of the total PAHs.

polycyclic aromatic hydrocarbons / grass / soil / mobile sources

Résumé – Évaluation de la contamination en hydrocarbures aromatiques polycycliques (HAP) de l'herbe et du sol dans les parcelles agricoles situées à proximité d'une autoroute et d'un aéroport. Le long des autoroutes et des aéroports, les HAP produits par les rejets de véhicules peuvent se déposer sur l'herbe et le sol et entraîner des risques de contamination de la chaîne alimentaire via le ruminant. De l'herbe et du sol ont été prélevés au plus près du bord de la route (10 m) et de la piste de décollage (50 m). Les analyses ont détecté la présence des 16 HAP listés par l'agence américaine de la protection de l'environnement à la fois sur les deux sites de prélèvement et dans l'herbe et le sol. Le site de l'autoroute est environ quatre fois plus contaminé (188 ng/g DW pour l'herbe) que le site de l'aéroport (69 ng/g DW) et des composés cancérogènes sont présents aussi bien dans l'herbe que dans le sol. De plus, la part des composés à faible poids moléculaire et à quatre et moins de quatre cycles est plus élevée dans l'herbe (36 %) que dans le sol où les composés à haut poids moléculaire et à plus de quatre cycles aromatiques représentent 86 % de la somme des HAP.

hydrocarbures aromatiques polycycliques / herbe / sol / sources mobiles

1. INTRODUCTION

PAHs are potentially mutagenic compounds, ubiquitous in the environment, produced by incomplete combustion occurring in natural events (volcanic eruption and forest fires...) and mainly during anthropogenic activities. Different kinds of pollution sources exist: stationary sources (coke plants, steel-works, incinerators and home heating...) and mobile sources (vehicle exhausts) [2]. The vicinity of roads and airports is known to be exposed to gasoline, diesel and kerosene vehicle exhausts (cars, trucks or aircraft) [1, 6, 7, 8, 14].

Motorways run along fields located directly behind the verge of the road or the motorway fence. In airport enclosures, many areas are still exploited by farmers. As vehicle exhausts

are considered to be one of the sources of PAHs, contamination of fields could be a major issue for food safety. It also seems necessary to assess animals' exposure through ingestion of fodder produced in agricultural plots near airports and motorways. A previous study has shown that organic compounds (such as PAHs) can be stored in the organism [3], leading to a potential risk of transfer through the food chain.

In this study PAH contamination levels by mobile sources are studied through their profiles found in grass and soil in the vicinity of a motorway and an airport located in the east part of France. Indeed, it is known that if ruminants ingest mainly grass, grazed directly in fields in summer, they also ingest soil (0.5% to 18% of the total ingested dry matter) [5].

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2. MATERIALS AND METHODS

2.1. Sampling site

In order to assess PAH contamination levels in grass and soil, samples were taken in the nearest fields along a motorway and an airport. We chose a motorway located in the North-East of France, directed North-South, connecting Luxembourg to the East of France. Its daily traffic is estimated to be about 70 000 vehicles. We sampled in a field located directly along the road, where ambient air measurements are regularly performed (except PAHs) and whose results characterise the place as a motorway air without any influence of other major sources of pollution.

The airport site is also located in the East of France, characterised by an international traffic of 112 000 planes per year. Samples were taken in a field located in the airport enclosure.

2.2. Sampling method

Five sub-samples each of grass and soil were collected in October along the perimeter of a 5-metre circle. The motorway sampling circle was located just behind the fence i.e. 10 metres from the road verge. The airport circle was located 50 metres from the end of the take-off strip.

Grass, mainly composed of *Poaceae* of eight months' vegetation, was cut with pre-washed scissors (using distilled water, acetone and hexane) 3 cm from the plant base to avoid collecting roots and soil. Each sub-sample was wrapped in an aluminium sheet then they were pooled together at the laboratory in order to obtain one representative sample. Grass samples were stocked immediately in a climate chamber regulated at 18–21 °C and 60%–80% hygrometry for five days. Dried samples were then ground into 1.5-mm pieces to perform PAH analysis.

The superficial parts of the soil (0–3 cm) were collected using a pre-washed garden dibble (using distilled water, acetone and hexane). The roots and grass parts were removed and sub-samples were pooled together in an aluminium container and placed in the same climate chamber as the grass. Soil samples were then ground and sieved through a 2-mm sieve.

Grass and soil samples were stored in solvent-washed glass pots to perform PAH analysis.

2.3. PAH analysis

Detailed extraction procedures are described in the NF ISO 15302 method, and were conducted by Micropolluant Technologies Ltd. PAHs were quantified by gas chromatography coupled to mass spectrometry (HP) in the single ion monitoring mode using internal deuterated standards: naphthalene- d_8 (m/z 136) for naphthalene; acenaphthene- d_{10} (m/z 164) for acenaphthene, acenaphthylene and fluorene; phenanthrene- d_{10} (m/z 188) for phenanthrene, anthracene, fluoranthene and pyrene; chrysene- d_{12} (m/z 240) for chrysene and benzo(a)anthracene and perylene- d_{12} (m/z 264) for perylene, benzo(b)fluoranthene, benzo(k)fluoranthene and benzo(a)pyrene.

3. RESULTS AND DISCUSSION

PAH analysis performed on grass and soil samples from both sites (airport and motorway) resulted in the detection of 16 PAHs listed by the US Environmental Protection Agency (Tab. I). Airport samples were less contaminated than motorway samples. Compounds considered as carcinogenic according to the US EPA – (benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, benzo(g,h,i)perylene and indeno(1,2,3-c-d)pyrene) – are present in all types of samples in different proportions. The ratio Indeno(1,2,3-c-d)pyrene/Benzo(g,h,i)perylene is about 0.9 for soil and 1 for grass by the motorway, suggesting mainly diesel vehicular emission at this sampling site [9].

3.1. PAHs in grass by the motorway and airport

The major PAHs in grass are phenanthrene and fluoranthene for both sites (Tab. I, Fig. 1). This result can be related to Bryselbout et al. study [4] along a motorway slope. The part consisting of carcinogenic compounds is also similar for both airport and motorway grass, respectively, 31% and 34%. Benzo(a)pyrene is detected in very low proportions: 1.5% for the airport and 3.4% for the motorway, but is six times more concentrated in motorway grass. Phenanthrene and fluoranthene are low toxicity molecules (TEF 0.001 for both compounds) of low molecular weight composed of only 3 and 4 rings.

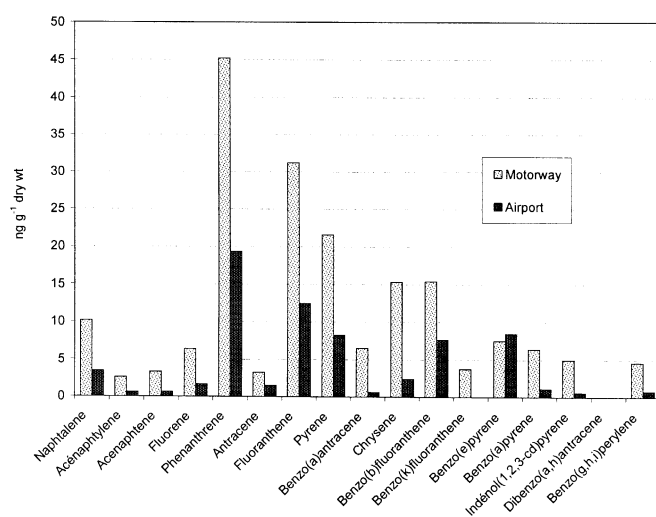
Total PAHs is different in the two sampling sites: 69 ng/g dry weight for the airport and 188 ng/g dry weight for the motorway. However, this last result is lower than those previously found in the vicinity of roads and motorways: from 1200 to 1461 ng/g dw in a similar matrix [1, 4, 14]. The differences in our results may come from the season of sampling: it is known that PAH concentrations range from a factor 1 to 10 between summer and winter [10], depending on the motorway traffic, the characteristics of vehicle emissions, the climate and the characteristics of surface leaves.

The differences in concentrations of PAHs between the airport and motorway range from a factor of 2.5 (phenanthrene, fluoranthene and pyrene) to a factor of 10 (Benzo(a)anthracene) and may be explained by the traffic and by emission production. Indeed, in this study the planes/vehicles ratio is about 1/230 and air analysis performed by airports show little incidence of PAHs from plane exhausts on air quality [11], and no typical PAH production [15].

PAH deposition on grass also depends on the gas-particle partition in the ambient air. Phenanthrene and fluoranthene (major PAHs in the samples) are compounds mostly found in the vapour phase in the air. Grass samples show few compounds found in the particle phase (Benzo(a)pyrene, Benzo(ghi)perylene, Benzo(a)anthracene and Chrysene) [10]. This suggests that gas phase PAHs could be adsorbed on leaves while particle phase PAHs could be washed off or windblown. Phenanthrene and fluoranthene are also low molecular weight compounds, easily transported and deposited on grass. Grass contamination could depend on air gas-particle partition that is linked to the climate conditions but also to the PAH profiles of the mobile sources studied. In

Table I. PAH concentrations by an airport and motorway (n = 2).

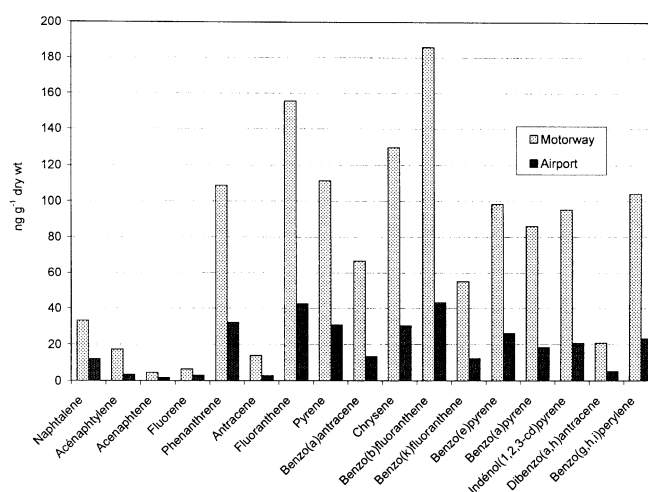
	Grass				Soil			
	Motorway		Airport		Motorway		Airport	
	(ng/g dry wt)	%	(ng/g dry wt)	%	(ng/g dry wt)	%	(ng/g dry wt)	%
Naphtalene	10.12	5.37	3.45	4.97	33.11	2.56	12.02	3.79
Acenaphtylene	2.59	1.37	0.59	0.85	17.32	1.33	3.41	1.06
Acenaphtene	3.34	1.77	0.62	0.89	4.6	0.35	1.68	0.52
Fluorene	6.38	3.39	1.64	2.36	6.56	0.5	3.01	0.93
Phenanthrene	45.18	24.01	19.31	27.87	108.8	8.41	32.09	9.98
Antracene	3.26	1.73	1.5	2.16	13.91	1.07	2.83	0.88
Fluoranthene	31.23	16.59	12.47	17.99	155.5	12.02	42.54	13.23
Pyrene	21.57	11.46	8.24	11.89	111.3	8.6	30.71	9.55
Benzo(a)antracene	6.5	3.45	0.63	0.9	66.64	5.15	13.44	4.18
Chrysene	15.33	8.14	2.38	3.43	129.8	10.03	30.37	9.44
Benzo(b)fluoranthene	15.41	8.19	7.61	10.98	185.8	14.37	43.17	13.42
Benzo(k)fluoranthene	3.74	1.98	n.d	n.d	55.04	4.25	12.34	3.83
Benzo(e)pyrene	7.5	3.98	8.46	12.21	98.31	7.6	26.12	8.12
Benzo(a)pyrene	6.41	3.4	1.06	1.53	86.07	6.65	18.42	5.73
Indénol(1,2,3-cd)pyrene	4.98	2.64	0.56	0.8	95.25	7.36	20.8	6.47
Dibenzo(a,h)antracene	n.d	n.d	n.d	n.d	20.8	1.6	5.21	1.62
Benzo(g,h,i)perylene	4.6	2.44	0.76	1.09	104.1	8.05	23.3	7.24
Σ PAH	188.14		69.28		1292.91		321.46	

**Figure 1.** PAH profile in grass collected near a motorway and airport (n = 2).

October (the month of our sampling), major contaminations could effectively come from vehicular emissions for the motorway sampling site, because phenanthrene and fluoranthene are part of vehicle exhaust emissions [1, 9].

3.2. PAHs in soil by the motorway and airport

The major PAHs found in soils are benzo(b)fluoranthene and fluoranthene for both the airport and motorway (Tab. I, Fig. 2). These compounds can be considered as typical of temperate soil [17], fluoranthene being more systematically detected [13, 16].

**Figure 2.** PAH profile in soil collected near a motorway and airport (n = 2).

The part consisting of carcinogenic compounds is almost the same for both the motorway and airport, respectively, 63% and 58%. Benzo(a)pyrene is detected in the same proportions in both sampling sites (6%). The detected PAHs are mainly of high molecular weight: 86% of the total PAHs are 4 and more than 4 ring compounds. This result can be related to a previous study performed in the UK [10].

Total PAHs is, respectively, 325 ng/g dry wt for the airport and 1292 ng/g dry wt for the motorway. Few references exist for airport soils and this result is similar to agricultural soil in Poland [12], suggesting low contamination levels at this site.

Table II. PAH properties in soil and grass.

	≥ 4 rings	< 4 rings
Soil	86%	14%
Grass	64%	36%

The differences in PAH concentrations between the airport and motorway soils range from a factor 3 to a factor 5. A motorway an airport do not have similar profiles and we have discussed above the main differences of the sampling sites. PAH concentrations in soil are due to the heaviest compounds mainly in the particle phase in ambient air, and contamination occurs by deposition on the surface soil. These PAHs also have a longer half-life and on the sampling sites they were under a continuous grass surface, protecting them from wind and photodegradation.

3.3. Soil and grass sample comparison

The contamination level of motorway samples being three times higher than airport samples for grass and four times higher than airport samples for soil, comparison between the grass and soil will be discussed using only the motorway results.

Fluoranthene is the only compound detected as a major PAH in both the grass and soil profiles. Two differences exist between these two profiles: the part consisting of carcinogenic compounds is more than 50% in soil, and total PAHs in soil is six times higher. As the samples of soil and grass were collected in the same place, accumulation is suggested in the soil. Grass cannot be considered as a filter above soil and a time effect could be suggested, explaining the higher concentrations in soils. Indeed, PAHs detected on grass could have been massively photodegraded in the previous summer months and the *poacea* lifetime is limited seasonally while degradation in soils depends on other factors (bacteria, organic matter, etc.) and accumulation is possible.

The soil and grass profiles show that soil is more concentrated in high molecular weight PAHs: 86% of compounds are of four and more than four rings, while they only represent 64% in grass (Tab. II). High molecular weight PAHs are also low volatility compounds and particle phase compounds found in the air (except for fluoranthene). Furthermore, these compounds have a longer half-life (17 000 h) in soil, suggesting that they are not rapidly degraded, unlike low molecular weight PAHs (5000 h).

Lower levels of grass contamination are due to lower concentrations of 4, 5 and 6 ring PAHs. This can be linked with the deposition properties of the compounds: the major PAHs of grass are mostly found in the gas phase, able to adsorb on leaves, while other PAHs can fall onto the soil. Higher levels of soil contamination are due to higher concentrations of more than 4 ring PAHs which are mainly in the particle phase and only deposited on leaves, easily washed off or windblown, or directly deposited on soil.

4. CONCLUSION

Grass and soil can be contaminated by PAHs when they are located by a motorway or an airport. Furthermore, they can be

ingested by ruminants. Even if the results show lower concentrations than other studies performed in other countries, the part consisting of carcinogenic compounds is up to 30% of the samples and the motorway is almost four times more contaminated than the airport.

In order to confirm these first results, other samples must be collected by other motorways and at different distances from the verge of the road to assess more precisely the real impact of vehicle exhausts on agricultural fields.

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