

Sand to Root Transfer of PAHs and PCBs by Carrots Grown on Sand with Pure Substances and Biosolids Amended Sand

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Keywords: biosolids, trace organic compounds, carrot, aquiculture, uptake

Abstract

A study on behaviour of trace organic compounds (Polycyclic Aromatic Hydrocarbons, PAH, and Polychlorinated Biphenyls, PCB) in a sand-plant system has been carried out, with the reclamation of wastewater treatment plant biosolids for agriculture in mind. Carrot plants (*Daucus carota*) were grown on soilless culture (sand), to provide optimal transfer conditions, in plant containers inside a temperature regulated greenhouse. There were two types of experiment. The trace organic compounds have initially been introduced as pure substances. A second experiment has been carried out under the same conditions, but using biosolids. Plant development has been unaffected by the presence of the pure substances and the biosolids. The transfer of the trace organic compounds has been followed in the peel, the core and the leaves of the carrot plants. Results obtained are expressed as fluxes of the trace organic compounds into the plant. The results clearly show that trace organic compounds accumulate in the carrot peel.

INTRODUCTION

In Europe, 40% of the annual production of biosolids from wastewater treatment plants is recycled in agriculture (Ademe, 1996). New Water Framework Directive (2000/60/EC) is currently under examination to standardise acceptable levels of trace organic compounds (TOC) in biosolids. It would appear essential, in the interests of public health, to study the transfer potential into plants of these TOC's (Diercxsens et al, 1987).

According to the French ministerial order of 8 January 1998, the maximum quantity authorized for land application of biosolids is 30 tons dry matter per ha per 10 years.

Carrots (*Daucus carota* L.) have been reported to be the crop having the greatest potential for organic uptake due to their high lipid content (Wild and Jones, 1992). In addition, carrots can be separated easily in peel, core and leaves in order to follow organic compounds transfer. Sand has no organic matter content thus applied organic compounds may be more bioavailable. So a root crop growing in sand can be considered the worst-case scenario to study behaviour of trace organic compounds.

The aim of this study was to investigate the transfer of Polycyclic Aromatic Hydrocarbons (PAHs) and Polychlorinated Biphenyls (PCBs) compounds from biosolids amended sand into a food-chain crop under "worst-case" operational practice (Morard, 1995).

MATERIALS AND METHODS

Fifteen days after germination, seedlings of carrots cv. Amsterdam A.B.K Bejo were transplanted into glass pots containing sand only (check, C₁), sand with PAH or PCB as pure substances (i.e. pure substance experiment) and containing sand only (check,

C₂) and sand with biosolids (check, B) (i.e. biosolids experiment, which started one year later). Four pots (2 L volume ; 15 10⁻² m high ; 13 10⁻² m diameter), each containing 7 carrots, per treatment were used.

For the pure substances experiment, seven PCBs isomers (28, 52, 101, 118, 138, 153 and 180 (IUPAC)) and the 16 PAHs of USEPA (Naphthalene, Acenaphthene, Acenaphthylene, Fluorene, Anthracene, Phenanthrene, Fluoranthene, Pyrene, Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Indéno[1,2,3-cd]pyrene, Benzo(ghi)perylene, Dibenzo(a,h)anthracene) were applied. According to the French ministerial order of 8 January 1998, the maximum quantity authorized for land application of biosolids is 30 tons dry matter per hectare per 10 years. For the pure substance experiment, the levels have been calculated from this maximum dose. For the biosolids experiment, the amount 30 tons dry matter of the biosolids studied per hectare were added to the pots in a single application.

Pots were randomly arranged in the greenhouse (mean temperature 24°C, 14 hours light, 50% relative humidity). All plants are watered with nutrient solution (KNO₃ 5, KH₂PO₄ 2, Ca(NO₃)₂ 5, MgSO₄ 1.5 mmol/L for macronutrients, and Fe 15, Mn 0.49, Cu 0.06, Zn 0.11, Mo 0.01, B 0.26 mg/L for micronutrient) .The liquid contained in the cups under the pots is poured back onto the sand. Carrots were harvested after 75 days and divided into leaves, peel and core.

The freeze-dried samples were extracted with hexane (Suprasolv, VWR Merck) for 3 hours with a Soxtec apparatus (Tecator 128). After a step of purification and concentration, the extract is analysed on a gas phase chromatograph coupled to a mass spectrometer detector (GC-MS) (Finnigan Trace 2000 Series) using the internal standard quantification method(Kömp et al., 2000 ; Müller et al, 2001 ; Sablayrolles et al., 2003).

Each extract has been analyzed twice, giving a total of 208 analyses. The control plants have been used as blank values for the analyses. The GC-MS apparatus gives concentration results in µg/L. Knowing the mass of the sample and taking into account all the analytical step, the results can be expressed in terms of µg/kg of dry matter.

The percentage transfer in the carrot (per gram dry matter) can be defined as :

$$\% \text{ transfer} = \frac{\text{mass of trace organic compounds per gram dry matter in carrot}}{\text{mass of trace organic compounds initially present in the pots}}$$

Analysis of variance of data and a Newman-Keuls multiple range test at P = 0.05 were performed.

RESULTS AND DISCUSSION

Newman-Keuls tests shows that there were no significant differences in growth between the carrots on the sand only (C₁) and those on the sand with pure substances (PAH and PCB) and between the carrots on the sand only (C₂) on the sand with the biosolids (B) (Table 1). The biosolids behaves as a nutrient solution prepared for ensure optimum plant development with no nutrition deficiency risk.

There was a large build up of PCBs and PAHs in the peel of the carrot which is thus acting as a barrier, stopping this group of compounds from moving into the root core (Table 2 and 3). The leaves and the core of the carrot seem unaffected by the addition of trace organic compounds. This observation has been confirmed by the repartition of trace organic compounds (Table 4) : mean of 74% in the peel, 17% in the core and 9% in the leaves. The fact that the PCB's and PAH's are confined to the epidermis is due to the characteristics of this particular plant. The carrot has large amounts of lipids and oil

channels favouring the uptake of non-polar compounds (Wild and Jones, 1992 ; Kipopoulou et al, 1999).

Whatever the type of experiment, the transfer percentages per gram of carrot were very low (Table 4). In the experiment using pure substances, the transfer of the PAHs compounds was less than 0.1% and the transfer of the PCBs compounds was around 0.8%. In the experiment using biosolids, the transfer of the PAHs compounds was 2.3% and the transfer of the PCBs compounds was 8.3%. The presence of surfactant compounds (e.g. laurylalkylbenzene sulfonates) in biosolids could explain the increase of transfer in the biosolids experiment.

CONCLUSIONS

There were no significant differences in growth between the carrots in the sand only, those in the sand plus pure substances and in the sand with the biosolids, and this was true for all the trace organics studied in the plant pots. Analysis of the carrot samples shows that the various members of the PCBs and PAHs families are mainly concentrated in the peel that is generally removed prior to consumption. This experiment demonstrates that the PCBs and PAHs compounds added to the soilless growth medium are found in part in the plants, but the percentage transfer remains low. Further researches should answer to following questions :

- How does this transfer percentage evolve as a function of the initial quantity of trace organic compounds in the growth medium ? Is this active or passive transport?
- Is the transfer of trace organic compounds facilitated when the latter are bound to the organic matter of the biosolids ?

Moreover, in the knowledge that European Regulations forbid any market gardening land application it will be necessary, in order to evaluate any real environmental impact, to study this transfer in crops grown on soil authorised to receive this type of amendment.

Literature cited

- ADEME. 2001. Les boues d'épuration municipales et leur utilisation en agriculture. Dossier Documentaire. 58 pp. Angers.
- Diercxsens, P., Wegmann, M., Daniel, R., Haeni, H., Tarradellas, J. 1987. Apport par les boues d'épuration de micropolluants organiques dans les sols et les cultures. *Gaz, eaux, eaux usées*. 3:123-132.
- Kipopoulou, A.M.; Manoli, E.; Samara, C. 1999. Bioconcentration of polycyclic aromatic hydrocarbons in vegetables grown in an industrial area. *Environmental Pollution*. 106:369-380.
- Kömp, P. and Lachlan, M.S. 2000. The kinetics and reversibility of partitioning of polychlorinated biphenyls between air and ryegrass, *The Sc. of the Tot. Env.*, 250 :63-71.
- Morard, P. 1995. Les cultures végétales hors sol. UPS. publications agricoles, Agen.
- Müller, J. F., D. W. Hawker, M. S. McLachlan, and D. W. Connel. 2001. PAHs, PCDD/DFs, HCB in leaves from Brisbane, Australia. *Chemosphere*, 43:507-515.
- Sablayrolles, C., Montréjaud-Vignoles , M., Treilhou, M., Benanou, D. and Patria, L. 2003. Optimal methods for analysing Polycyclic Aromatic Hydrocarbons and Polychlorobiphenyls content of carrots as vegetal matrix, 4th European Meeting on Environmental Chemistry, Plymouth, UK 10-13 December, p 116.
- Wild, S.R. and Jones, K.C. 1992. Organic chemicals in the environment: polynuclear aromatic hydrocarbon uptake by carrots grown in biosolids-amended soil. *J. Environ. Qual.* 21:217-225.

Tables

Table 1. Dry matter production (g per plant, mean \pm SE)

Pure substance experiments				Biosolids experiments			
Treatment	Peel	Core	Leaves	Treatment	Peel	Core	Leaves
C ₁	0.67 \pm 0.05 a	1.70 \pm 0.21 a	0.82 \pm 0.08 a	C ₂	0.47 \pm 0.03 a	0.95 \pm 0.10 a	0.35 \pm 0.20 a
PAH	0.57 \pm 0.06 a	1.53 \pm 0.21 a	0.93 \pm 0.08 a	B	0.63 \pm 0.30 a	0.63 \pm 0.10 a	0.82 \pm 0.31 a
PCB	0.51 \pm 0.04 a	1.19 \pm 0.13 a	0.63 \pm 0.04 a				

Mean values of four replications followed by the same letter in a column are not significantly different at $P < 0.05$; \pm SE, standard error in variance analysis.

Table 2. Total content ($\mu\text{g}/\text{kg}$ DM, mean \pm SE) of PAH and PCB in sand and in the different parts of carrot plants in the pure substance experiment.

Treatment	Sand	Peel	Core	Leaves
Σ PAH	667 \pm 60	3071 \pm 161	345 \pm 18	151 \pm 6
Σ PCB	107 \pm 9	11995 \pm 451	316 \pm 14	840 \pm 47

Table 3. Total content ($\mu\text{g}/\text{kg}$ DM, mean \pm SE) of PAH and PCB in sand and in the different parts of carrot plants in the biosolids experiment.

Treatment	Sand	Peel	Core	Leaves
Σ PAH	2.60 \pm 0.22	549 \pm 37	107 \pm 9	27 \pm 1
Σ PCB	0.20 \pm 0.02	189 \pm 12	20 \pm 2	19 \pm 1

Table 4. % of transfer (per gram dry matter of carrot) of Σ PAH and Σ PCB and % distribution of Σ PAH and Σ PCB transferred in the different parts of carrot plants

	Pure substance experiments		Biosolids experiments	
	% transfer	% repartition	% transfer	% repartition
Σ PAH	< 0.1	{ 74% in peel 22% in core 4% in leaves	2.3 \pm 0.5	{ 65% in peel 30% in core 5% in leaves
Σ PCB	0.8 \pm 0.1	{ 85% in peel 5% in core 10% in leaves	8.3 \pm 1.7	{ 70% in peel 12% in core 18% in leaves