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Treatment of tunnel wash waters – experiments with organic sorbent materials. Part I: Removal of polycyclic aromatic hydrocarbons and nonpolar oil

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Abstract

Tunnel wash waters characterize all waters that run off after washing procedures of tunnels are performed. These waters represent a wide spectrum of organic and inorganic pollutants, such as polycyclic aromatic hydrocarbons (PAHs) and toxic metals. Removal of such contaminants from water runoff was investigated using laboratory tests after washing procedure was performed on two road tunnels in eastern Norway (Hanekleiv and Bragernes). Due to diverse character of both, treatment media and treated wash waters, the whole investigation was divided into two separate laboratory experiments. The treatment efficiencies were established based on the levels of concentrations and reductions of the measured contaminants in the effluents released from the tested media. In the first part of the article, the contents of nonpolar oil (NPO), 16 individual PAHs, and total PAHs are described. This part revealed that the combination of two organic sorbent materials provided the highest treatment efficiency for wash waters released from the road tunnel and from electrostatic filters. The greatest reduction levels reached 97.6% for NPO, 97.2% for benzo[a]pyrene, and 96.5% for the total PAHs. In the second part of the article, the concentrations and the removal rates of toxic metals are reported.

Key words: electrostatic filters; nonpolar oil (NPO); organic sorbent materials; polycyclic aromatic hydrocarbons (PAHs); tunnel; wash water

Introduction

The majority of studies on water pollution resulting from road runoff mainly focused on the characterization of quality and quantity of stormwater. However, it represents only "open" road areas, directly affected by external weather conditions. Therefore, an impact of pollution from sources other than vehicular traffic, in particular, exposure to urban and industrial airborne pollutants, cannot be directly excluded. However, the effect of some parts of roadways may be "isolated" from that of the external factors and thus can represent the substantial impact of automobile traffic on generated pollution. This, in particular, refers to road tunnels that constituted specific, "closed" section of road areas, which varied from several meters up to several tens of km, the world's longest road tunnel, 24,510 m (Laerdal, Norway).

Studies carried out by Pereira *et al.* (2002) and Wang *et al.* (2005) showed that high concentrations of organic compounds and trace metals are strongly related to road tunnel areas. Chellam *et al.* (2005) investigated fine particulate matter in a highway tunnel (Houston, USA) and described high correlation of increasing concentrations of some or-

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ganic compounds with percentage carbon emanating from diesel vehicles. However, a study conducted by Manoli *et al.* (2004) revealed that the highest content of polycyclic content of polycyclic aromatic hydrocarbons (PAHs) was emitted from gasoline engines compared with diesels.

Fine particles emitted from vehicles have adverse health effect (Lin et al., 2005) and PAHs have carcinogenic and mutagenic potency (Durant et al., 1996; Hannigan et al., 1998; Nielsen et al., 1996). Van Dolah et al. (2005) reported that total concentration of PAHs in sediments adjacent to roads decrease with increasing distance from the roads. In addition, study on transport of traffic-related aerosols revealed that the contribution of traffic to the fine particle concentration could account for up to 70% in the closest vicinity of the roads (Wrobel et al., 2000). The relationships between the concentrations of compounds and their transport distances may indicate that most of the traffic pollutants are accumulated in road tunnels. This relates not only to the road surfaces but also to the walls, roofs, and electrostatic filters as well as all other internal devices of the tunnel. Although part of these pollutants may be periodically washed out (mainly from roads) by some water flowing out from tunnels, the greatest quantity of them can only be removed during the cleaning/maintenance services that are performed inside



tunnels.

Water that runs off after washing of road tunnels and their electrostatic filters contains a complex mix of pollutants, including PAHs, oil, detergents, and easily degradable organic matter (Roseth *et al.*, 2003). The bioavailability of these pollutants raises due to the use of different soap components for tunnel maintenance (Andersen and Vethe, 1994). Various types of cleaning detergents are introduced to remove adhesive and oily pollutant components from the road tunnels, but even those "environmentally friendly" and easily degradable detergents have acute toxic effects on aquatic organisms before they are degraded.

Toxicity of wash water, the level of toxicity being similar to urban stormwater and road runoff (Boxall and Maltby, 1997; Pitt *et al.*, 1995), is mainly related to the same group of inorganic and organic toxicants. Most of the components of pollutants in wash water released from both, road tunnels and tunnel electrostatic filters, are attached to particles and therefore may be removed in sedimentation tanks or ponds. However, some pollutants can still pass through sedimentation traps (Farm, 2002; Lundberg *et al.*, 1999), and some of these, e.g., bioavailable components, even in small concentrations, can potentially cause toxic effects on aquatic fauna and flora (Boxall and Maltby, 1995). The efficient treatment of wash water is therefore necessary before its discharge into the closest stream or sewer pipeline.

Since our previous investigations have demonstrated that organic media based on bark, cellulose, and peat could remove "not particle attached" harmful organic and inorganic compounds from polluted runoffs, this research was mainly undertaken to evaluate four selected, commercially available, sorbent materials, and to assess their efficiencies in the removal of "not particle attached" pollutants from presedimented wash waters that run off from road tunnel and electrostatic filters. Therefore, the overall aim of the research focused mainly on the practical aspects of using organic sorbents for treatment of these kinds of waters and thus sorption isotherms, kinetics, mechanisms, and capacity determined from the tested sorbents were not considered.

The selection of the sorbent materials was made based on the revealed sorption properties of these media and on the removal of large amounts of organic and inorganic pollutants from contaminated waters, stormwaters, and wastewaters.

1 Materials and methods

1.1 Study sites

Due to the diverse character of tested materials, the whole investigation was performed in two independent laboratory experiments. The first one, column experiment, focused mainly on treatment of wash water from road tunnel using columns filled in with organic sorbent materials. The second one, bag experiment, determined the efficiency of filtration sacks treating wash water from electrostatic filters of road tunnel. The laboratory-scale studies were carried out at Bioforsk Soil and Environment (Aas, Norway).

1.2 Column experiment

The efficiency of treatment of wash water from roof, walls, and road surface of motorway tunnel, Hanekleiv (1.8 km, Southeast Norway), was studied during a 1-month experiment.

The PVC pipe columns, having approximately the same size (7 cm diameter and 40 cm length) and volume (1.5 L) were filled in with four organic sorbent materials based on bark, cellulose, and peat. The sorbents were characterized by A, Pm, Po, and M (Table 1). In addition, an experimental combination of two sorbents, Po and M, was also tested. The combination Po-M was made not as a random mixture of these two materials but as a successive filling of the column with two separate layers of Po and M. The first half of the column (20 cm from the bottom) was filled in with Po, whereas the second half (20 cm up to the top) contained M. One column was established as a background for others and was run as blank during the entire experiment.

To deliver approximately equal doses of wash water to the columns, a peristaltic pump with adjusted PVC hoses, connected to the bottom (inlet section) of each of the columns, was used. Application of wash water was calculated for the mean hydraulic loading rate (HLR) of the tested sorbent media that is supposed to occur under the real conditions, i.e., average production of wash water during cleaning procedure of road tunnels. The pump was set to operate continuously and to deliver a flow rate of 19-25 L/d (HLR of $5-7 \text{ m}^3/(\text{m}^2 \cdot \text{d})$).

After the media inside the columns had been saturated by an upflow of wash water, the effluents appeared in the outlet hoses, fixed on the top (outlet section) of each of the columns. Samples of these effluents were collected in glass

Acronym	Explanation	Description	Producer, distributor			
A	Axon	Commercially available peat-based material	Arcon AS (Axon Miljöteknik AB), Sweden			
Pm	PROLupBLÅ (mix)	Commercially available mixture of organic materials that possess properties for adsorption of oil and metal contaminants	Prolup Absorbenter AB, Sweden			
Ро	PROLupBLÅ (oil)	Commercially available organic material that possesses properties to adsorb oil contaminants	Prolup Absorbenter AB, Sweden			
М	Metal bark	Commercially available wet bark that possesses properties to adsorb metal contaminants	Stadspartner AB, Sweden			
Po-M	PROLupBLÅ (oil) and metal bark	Combination of two organic materials that was made for the purpose of the experiment	Prolup Absorbenter AB, Sweden and Stadspartner AB, Sweden			

 Table 1
 Description of organic sorbents

Several grab samples of untreated wash water were taken during the entire experiment, and based on them, a composite sample was prepared for chemical analyses.

1.3 Bag experiment

The treatment efficiency of wash water from electrostatic filters of road tunnel, Bragernes (2.3 km, Drammen, Norway), was studied by using commercial sack filters (Teknobag-Draimad) that are originally designed for dewatering and bagging sludge from water and effluent purification plants.

For the purpose of the study, the original width of the sacks was adjusted to 5 cm, to reduce both the mass of the organic media and the time of filtration. The reason for not changing the length of the sack was to retain filtration path for wash water through the originally designed long sack (118 cm). Consequently, the experimental bag had a volume of 2.3 L.

A pair of such bags was established for a study, where one of them was run as blank, the other one was filled in with organic media. The media constituted the same combination Po-M of two organic sorbent materials as in the column experiment. Moreover, the two separated layers of the combination Po-M also corresponded to the inlet and the outlet part of the bag. The first half of the bag (50 cm from the bottom) was filled in with M, whereas the second half (50 cm up to the top) contained Po. The whole experiment was established for a 1-d test and involved seven sampling sequences. Every one hour, 2 L of wash water was poured into the bags (HLR of $24 \text{ m}^3/(\text{m}^2 \cdot \text{d})$). The effluents from the bags were collected continuously into glass cylinders, from which two samples (each characterizing the effluent from the empty bag and from the bag with organic sorbents, respectively) were taken for chemical analyses.

1.4 Methods

All samples collected from columns and bags were analyzed for the same group of organic parameters, i.e., PAHs and NPO (nonpolar oil).

Determination of 16 USEPA-priority PAH substances and their total amount was performed according to the USEPA analytic technique, EPA 8100. The samples were analyzed using a gas chromatography and a mass spectrometry (GC/MS) using selected ion monitoring (SIM) with electron-ionization (EI) mode. The analyzed PAH compounds and their limits of quantitation (LOQ) are

 Table 2
 Limits of quantitation (LOQ) for the investigated polycyclic aromatic hydrocarbons (PAHs)

$LOQ~(\mu g/L)$	PAHs
0.01	Acenaphthylene; anthracene; benzo[a]anthracene;
	benzo[a]pyrene; benzo[b]fluoranthene;
	benzo[g,h,I]-perylene; benzo[k]fluoranthene;
	dibenzo[a,h]anthracene; indeno[1,2,3-cd]pyrene
0.05	Chrysene; fluoranthene; fluorene; phenanthrene; pyrene
0.10	Acenaphthene; naphthalene; total PAHs

presented in Table 2.

Concentration of NPO was determined in accordance with Method 1664 of the USEPA and with Standards for Water Quality ISO 9377-2. All samples were extracted with hexane and were analyzed on a gas chromatograph that was equipped with a flame-ionization detector (GC/FID).

2 Results and discussion

2.1 Column experiment

Concentrations of the investigated components in the effluents eluted from the columns filled in with the organic sorbent materials were compared with the respective concentrations in both background samples, i.e., the effluents eluted from the empty column (E) and those from the wash water of road tunnel (TW) applied during the experiment (Table 3).

Although half of the 16 priority PAHs analyzed in TW had concentrations below their LOQ, the obtained results showed that the general content of these organics corresponded with values of toxic pollutants in urban stormwaters (Makepeace *et al.*, 1995; Pitt *et al.*, 1995) and thus uncontrolled discharge of untreated tunnel wash waters may have toxic effects on aquatic life. This, in particular, refers to some toxicants, e.g., benzo[a]pyrene, fluoranthene, and others that are restricted by standards and criteria established for surface water quality.

Concentration of the most common carcinogenic PAH, benzo[a]pyrene, which was found in TW (Table 3), was found to be higher than the limited values classified by the European Commission (2006) in the Environmental Quality Standards (EQS) for priority substances in surface water. The concentration exceeded both values of benzo[a]pyrene defined as an annual average (AA-EQS) 0.05 µg/L and a maximum allowable concentration (MAC-EQS) 0.1 µg/L in inland and other surface waters. Content of this toxicant recorded in sample of TW was also higher than the value of 0.015 µg/L established by the Canadian Council of Ministers of the Environment (CCME) for the protection of freshwater aquatic life (Alberta Environment, 1999). In addition, concentration of benzo[a]pyrene in tested TW exceeded value of this pollutant (0.0038 µg/L) described in the USEPA (2006) National Recommended Water Quality Criteria for the protection of aquatic life and human health. However, the value obtained from the American criteria expresses, carcinogenicity criterion of 10^{-6} risk for human health when water with organisms are consumed.

Among other PAHs that were present in TW (Table 3), four additional parameters showed exceeding values of the limited concentrations established for surface water qualities. These involved indeno[1,2,3-cd]pyrene, fluoranthene, pyrene, and naphthalene. However, none of them had higher concentration of the effluents from the sorbents than their LOQ. As for the total PAHs, the highest concentration was recorded for the effluents eluted from the column that contained M (Table 3). In addition, this material produced effluents with the quantitative concentration, even when

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Table 3	Concentrations and maximal reductions of PAHs and NPO in the background samples, the empty column (E) and wash water of road tunnel
	(TW), and the effluents from the columns filled in with the organic sorbent materials (A, Pm, Po, M, and Po-M)

Parameter	TW	Initial sample (µg/L)						Final sample (µg/L)				Reduction*	
		Е	А	Pm	Ро	М	Po-M	А	Pm	Ро	М	Po-M	(%)
Indeno[1,2,3-cd]pyrene	0.08	0.01	< 0.01	0.01	< 0.01	0.01	< 0.01	< 0.01	< 0.01	0.01	0.01	< 0.01	87.5
Acenaphthylene	0.06	0.01	< 0.01	0.01	0.01	0.01	0.02	0.01	< 0.01	0.01	0.01	0.01	83.3
Fluorene	0.07	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	28.6
Phenanthrene	0.17	0.17	< 0.05	0.11	< 0.05	0.17	0.06	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	70.6
Benzo[a]pyrene	0.36	0.02	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	97.2
Fluoranthene	0.37	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	86.5
Pyrene	0.61	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	91.8
Naphthalene	1.15	< 0.10	< 0.10	0.13	0.17	0.47	0.59	< 0.10	< 0.10	< 0.10	< 0.10	< 0.10	91.3
Total PAHs	2.86	0.12	0.12	0.32	0.19	0.74	0.68	< 0.1	< 0.1	< 0.1	0.14	< 0.1	96.5
NPO	4,200	470	180	180	100	230	< 100	< 100	< 100	< 100	< 100	< 100	97.6

* Maximal reductions calculated with respect to LOQ.

effluents from other media showed concentrations of the total PAHs below LOQ.

Treatment of wash water from road tunnel by the organic sorbent materials revealed high reductions of PAHs, among which benzo[a]pyrene was reduced by 97.2% (Table 3). Its concentrations were lower than those in the effluents from eluted E and did not exceed LOQ of 0.01 μ g/L. Thus, benzo[a]pyrene contents were also lower than the limited concentrations defined by standards and guidelines for surface water quality (Alberta Environment, 1999; European Commission, 2006). Maximal reduction of the pollutants during the experimental period was attained for NPO 97.6% (Table 3). The reduction percentage was the same for all sorbents that exhibited properties to adsorb organic and inorganic pollutants.

2.2 Bag experiment

Contents of the investigated components in the effluent from the bag filled in with the sorbent materials (BF) were compared with contents of these organics in both background samples, i.e., the effluents from the empty bag (EB) and wash water of tunnel electrostatic filters (WF) applied during the experiment (Table 4).

The obtained results showed that only two of the 16 individual PAHs measured in WF had concentrations below their LOQ, i.e., dibenzo[a,h]anthracene < 0.01 μ g/L and acenaphthene < 0.10 μ g/L. In addition, the results revealed that WF contained more toxic pollutants, which exceeded the values set by standards and criteria established for surface water quality, than TW tested in the column experiment. That state was exemplified by concentration of 11 individual PAHs, among which six compounds (benzo[g,h,I]perylene, indeno[1,2,3-cd]pyrene, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene, and fluoranthene) showed higher values compared with the limited values of the EQS for priority substances in surface water (European Commission, 2006). Six PAHs (anthracene, benzo[a]pyrene, benzo[a]anthracene, phenanthrene, fluoranthene, and pyrene) in WF were also observed at concentrations greater than the respective concentrations of CCME for the protection of freshwater aquatic life (Alberta Environment, 1999). In addition, carcinogenicity criterion of the American Water Quality Criteria

for Priority Toxic Pollutants (USEPA, 2006) was also exceeded by six organics (indeno[1,2,3-cd]pyrene, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]yrene, chrysene, and benzo[a]anthracene) that were present in WF. Although most of these parameters in WF had values at the acceptable level of at least one of the surface water quality norms, benzo[a]pyrene concentration exceeded the limited values in waters defined by the European Commission (2006), CCME (Alberta Environment, 1999), and USEPA (2006).

The effluent from EB contained 10 individual PAHs at higher levels than the allowable concentrations for the protection of surface water quality. These parameters, except phenanthrene, overlapped the 11 excessive PAHs revealed in WF.

The lowest contents of almost all individual PAHs (except naphthalene) as well as the total PAHs characterized the effluent from BF (Table 4). In addition, concentrations of chrysene and fluorene in this effluent were lower than their LOQ of 0.05 µg/L. The noted exception revealed that the effluent from BF had the highest concentration of naphthalene compared with the values, which characterized both background samples, EB and WF. The reason for that was not only the process of leaching out of naphthalene from the sorbent combination Po-M, on the one hand, but also the leaching out of this PAH from the bag. On the other hand, because the lowest concentration of naphthalene was recorded in applied WF (Table 4). Nevertheless, its concentrations were still below the limited values defined by standards and guidelines for surface water quality (Alberta Environment, 1999; European Commission, 2006). The limited values were exceeded, however, by concentrations of eight parameters in the effluent from BF that overlapped the 10 excessive PAHs revealed in the effluent from EB. Two organics, anthracene and chrysene, were reduced to the allowable concentrations of toxic pollutants in surface waters. In addition, concentration of benzo[a]pyrene was lower than its limited values of the EQS (European Commission, 2006). The highest reductions among the individual PAHs were found for pyrene (80.6%), fluoranthene (80.3%), and anthracene (80.0%). The value of the total PAHs was reduced by 67.1% (Table 4).

The effluent from BF also revealed the lowest content

Parameter		Concentration (µg/L)	Reduction (%)		
	WF	EB	BF	WF-EB	WF-BF
Acenaphthylene	0.02	0.04	0.01	-100	50.0
Benzo[g,h,i]-perylene	0.04	0.03	0.01	25	75.0
Indeno[1,2,3-cd]pyrene	0.04	0.03	0.01	25	75.0
Anthracene	0.05	0.04	0.01	20	80.0
Benzo[k]fluoranthene	0.05	0.05	0.02	0	60.0
Benzo[a]pyrene	0.07	0.05	0.02	29	71.4
Fluorene	0.08	0.06	< 0.05	25	37.5*
Benzo[b]fluoranthene	0.11	0.08	0.03	27	72.7
Chrysene	0.14	0.09	< 0.05	36	64.3*
Benzo[a]anthracene	0.20	0.13	0.05	35	75.0
Naphthalene	0.29	0.32	0.37	-10	-27.6
Phenanthrene	0.49	0.32	0.11	35	77.6
Fluoranthene	0.71	0.49	0.14	31	80.3
Pyrene	0.98	0.66	0.19	33	80.6
Total PAHs	3.25	2.39	1.07	26	67.1
NPO	66,300	80,500	45,400	-21	31.5

 Table 4
 Concentrations and reductions of PAHs and NPO in wash water of tunnel electrostatic filters (WF), the effluents from the empty bag (EB), and the bag filled in with the sorbent materials (BF)

* Reduction calculated with respect to LOQ.

of NPO (Table 4). Higher value was recorded in applied WF; however, the maximum was found in the effluent from EB. This indicated that organic sorbent materials could not only remove NPO from applied wash water, but could also reduce the effect of leaching out from the bag. In this context, concentration of NPO was reduced by 31.5% (Table 4).

3 Conclusions

The spectrum of organic contaminants, which were present in wash waters from road tunnels and electrostatic filters, indicated that these waters require treatment procedures before direct discharge into any water body.

The obtained results from both experiments revealed that WF contained more toxic pollutants, which exceeded standards and criteria established for surface water quality, than common TW.

The column experiment showed that the tested sorbent materials effectively removed organic pollutants from TW. The lowest contents of PAHs were already observed at the beginning of the experiment, in the initial sample of the effluent from sorbent A. At the same time, the sorbent combination Po-M reduced NPO concentration to the level below LOQ. As for the final effluents, the samples collected from Pm, A, Po-M, and Po revealed the lowest concentrations of the measured organics, not higher than their LOQ.

The bag experiment showed that the most efficient treatment of WF was provided by BF. Thus, the lowest concentrations and the highest reduction levels of almost all individual PAHs, the total PAHs, and NPO were found in the effluents from the sorbent combination. Among all measured parameters, only concentration of naphthalene in the effluent from BF was higher than its concentrations in both background samples, i.e., WF and EB. That, however, was the effect of leaching out of naphthalene from the bag and from the tested sorbents.

Although the organic sorbents could effectively treat tunnel wash waters, further investigations are still needed for better understanding of the removal mechanisms occurring in the sorbent media. For this purpose, the performance of sorption tests would be recommended under representative conditions, different from those established in the experiments. Besides, to obtain equilibrium and relevant isotherms, sorbates in prepared background solution need to be used, instead of wash waters that represent complex sources of pollutants originating from combustion, traffic-related materials, road dust, and some water additives (such as soaps and detergents). In addition, these types of polluted waters have not been yet widely investigated and hence there is a lack of adequate research work and available published data. Moreover, extended studies are also recommended for establishing guidelines for designing and dimensioning of a full-scale system treating wash water runoffs from road tunnels.

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