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Enhanced Composting for Cold-Climate Biodegradation of Organic Contamination in Soil

by

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ABSTRACT

Bioremediation of soils contaminated with hazardous wastes is becoming a preferred technology because of its simplicity, lack of residuals requiring special handling, and relatively low cost when compared with traditional alternatives. Bioremediation was evaluated as an alternative for a coke works site in northern Norway near the Arctic circle, which was characterized in 1989 as having significant contamination by polycyclic aromatic hydrocarbons (PAH). About 20,000 tons of soil containing PAH's (ca. 500 mg/kg) were excavated. Groundwater at the site contained ca. 2-3 mg/l and 0.4-1.6 mg/l naphthalene and benzol. A pilot study was conducted in 1990, in which 1,000 m^3 of soil were treated in an enhanced composting system. Composting was chosen over landfarming or slurry reactors because of: low capital and operating costs, on-site capability (low area requirement), minimal developmental requirements, capability for cold-climate, year-round operation.

The variables tested were: N & P, bark matrix and dispersant addition, temperature (4°-16°C), moisture (10-35 %), and aeration by blowers, H_2O_2 addition or pile turning. Composite soil samples from five sampling points from each pile were taken twice weekly for PAH analyses (GC-MS) and soil moisture. Soil gas CO_2 and O_2, temperature, pH, and odor were measured onsite twice weekly. The treatment objective was ≤ 10 mg/kg Total PAH. Results showed that the PAH-content was reduced to below the objective within 8 weeks at 12-16°C. The "lag phase" for biodegradation was ca. 1 week under optimal conditions versus 8 weeks for unamended compost piles. Thereafter, degradation kinetics were biphasic with approximately 90 % PAH removal within 4 weeks under optimal conditions. Treatment efficiency ranged from 96-99 % dependent on test variables. Optimal results were obtained by 1) addition of tree bark as a matrix, 2) supplemental forced aeration, 3) soil moisture maintained at 25-30 % for this soil type, 4) N additives, and 5) dispersant additives. Provision for pile warming during winter operation by heating cables in the pads is desirable, although cultures were developed which performed satisfactorily at 4°C. The process includes soil sorting, mixing with tree bark and amendments (nutrients and dispersant). The soil is placed in rows ca. 2 m wide x 1.5 m high on geomembranes. Forced aeration is not initiated for 2-4 weeks until the more volatile aromatics (e.g. naphthalene) have been degraded. Treatment costs are estimated to be NOK 1350/ton or approx. $ 200/t. (Lower labor and material costs will likely

prevail outside of Scandinavia). The costs include pile-to-pile material handling, materials, operations, and analytical control.

INTRODUCTION

The remediation of a contaminated coke works site (Norsk Koksverk) i Northern Norway was initiated in 1989. It was unique in that it was Norway's first major cleanup in which several technologies had to be considered for the contaminants, including polycyclic aromatic hydrocarbons (PAH), arsenic, cyanide, and copper. The subject of this paper concerns the treatment of the PAH-contaminated soil, in which a biological process was chosen for the pilot study. The treatment of the other contamintants in both soil and groundwater have been described in reports in Norwegian and will be made available in English publications in the near future.

Biological treatment of soils contaminated with organics is a preferred technology in many cases because of its simplicity, lack of residuals (e.g. sludges) requiring further treatment, and relatively low cost. The technology for excavated soils is generally applied in three process types: land farming, composting or slurry reactors. In situ biorestoration is also applicable, circumstances permitting. All of these processes are in use internationally and have recently been reported by others (Sims, et al, 1989; Steps, 1989; Borow and Kinsella, 1989; Christiansen, et al, 1989).

Coke or gas works sites are typically contaminated by PAH's (Turney and Goerlitz, 1990), which are also components of many hydrocarbon products. Since these compounds are relatively refractory, carcenogenic, and bioaccumulate, there is considerable interest to effectively treat contaminated sites. PAH's can be biologically degraded by naturally occuring bacteria and fungi (Park, et al, 1990; Pothuluri, 1990) and adapted cultures (Portier, 1989). Bioremediation processes especially designed for aromatics have also recently been described (Mahaffy and Compeau, 1990; Bewley and Theile, 1988; Compeau, et al, 1990; Tan, et al, 1990; and Stroo, et al, 1989).

Composting technology was chosen for further investigation at pilot scale after having determined at bench scale that the contaminants were biodegradable. Composting was chosen over landfarming because of the better opportunity for temperature control in a rather cold climate, and because of smaller area requirements. Composting was chosen over slurry reactors because of simplicity and the lack of a substantial investment requirement in reactors and process controls. A description of the pilot study and the proposal for full scale remediation follows.

NORSK KOKSVERK SITE - NORWAY

History

The Norwegian Parliament (Stortinget) decided in 1961 to build a coke plant located in Mo i Rana in the northern part of Norway. The state owned steel mill (Norsk Jernverk A/S) who

would be the major user of the coke, was also located in Mo i Rana. The plant processed approximately 440 000 tons of coal per year. The coal was primarily shipped in from Spitsbergen.

The plant began operations in 1964. The ammonia production was started 6 months later. The plant annually produced 55-60 000 tons NH_4, approx. 15000 tons tar and 5000 tons benzene. These products were sold without further processing at the plant.

The area of the industrial site is 250 ha (101 acres). The various activities are shown in Figure 1. The plant was closed down in the fall of 1988 for economic reasons. Site characterization and clean-up was required by the pollution control authorities (SFT) before any further development of the area would be permitted.

Contamination at the site is the result of both routine operations over the 27 year plant lifetime plus a recent accident. The primary contaminant associated with operations involved intermittent leakage of benzene and other aromatic solvents to the site, resulting in a ca. 10-15 cm floating layer of aromatics on the groundwater; the second was a spill of an alkaline arsenic solution (ca. 200 m^3). Both involved surface tanks in two separate areas of the site. Preliminary analyses also revealed contamination by copper, cyanide, PAH, and other aromatics than benzene.

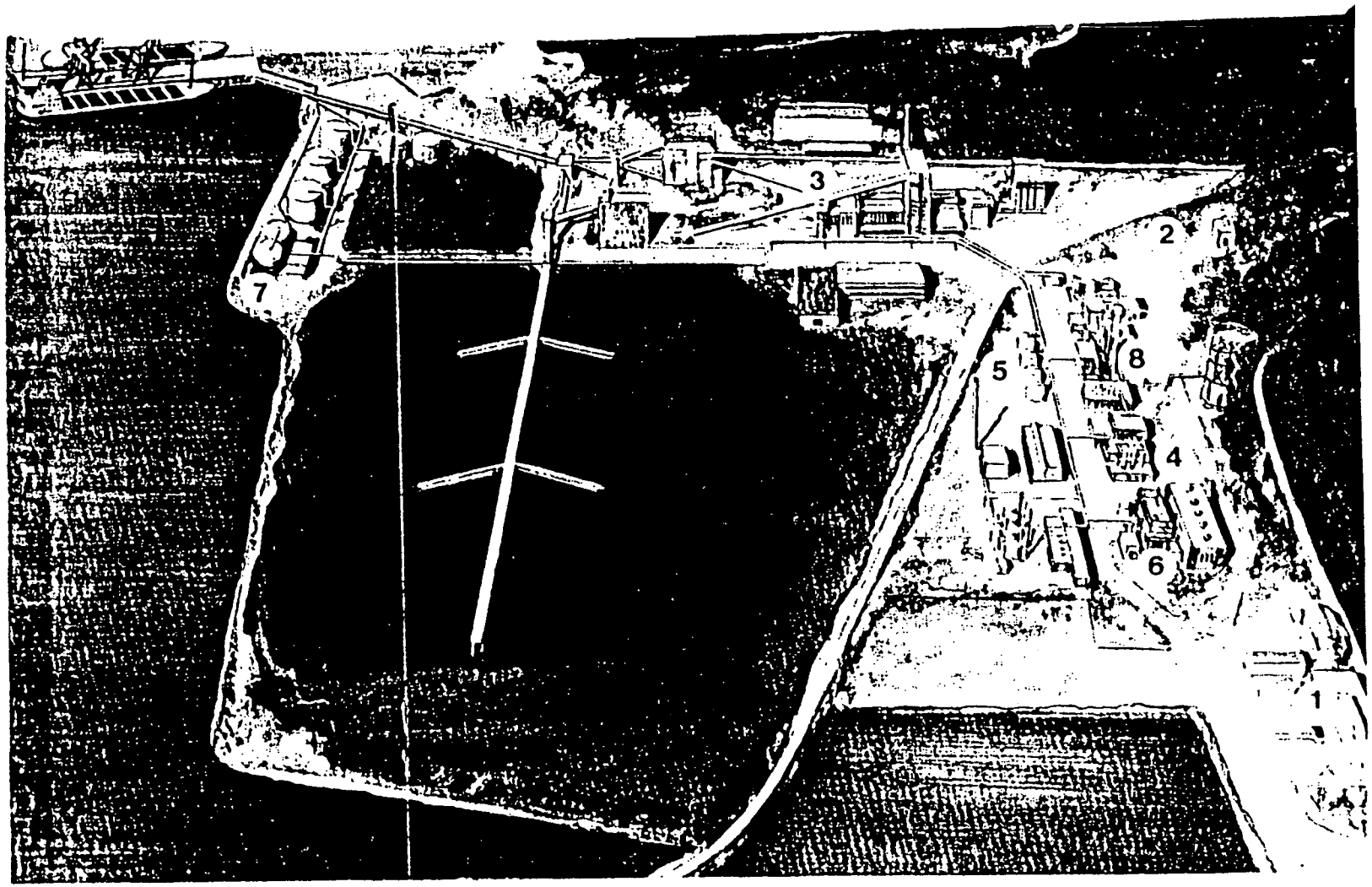
Site Characterization/Contamination Survey

To the north a rocky ridge extends to the west. The rockface can be seen in the open terrain. The natural sediments in the area consist of silt and clay. The clay content increases with depth. The site itself is sedimentary material or sandy fill from 2 to 3 m thick on a clay layer which is the original fjord bottom.

The railroad track divides the area in two. The lower portion of the area, outside the clean-up area, is filled in with bottom sediments from the fjord and slag from the steel mill. The slag has the same particle size as coarse sand.

The groundwater consists primarily of surface runoff that infiltrates the area. All groundwater movement is shallow and directed toward the fjord, which is the final recipient. Tidal changes influence the groundwater movement (variation - 1.7 m to + 1.3 m).

To identify the contamination in the area 45 test wells were eventually installed. Soil camples and groundwater samples were collected at each well. In addition to the test wells, georadar (SIR-3) was used to point out areas with concentrated deposits of waste, e.g. buried drums.



1 Administration
2 Laboratory
3 Coke oven
4 Benzen production
5 Benzen storage
6 Ammonia production
7 Ammonia storage
8 Sulphur treatment plant

Figure 1. Norsk Koksverk Plant in Mo i Rana, Norway

Figure 2 shows the location of the sampling wells and seven areas requiring remediation. Owing to the acute contamination of two areas, approx. 20,000 tons of soil from area "1" was excavated and placed in lined and covered depositories on the site (Dep. 1-4). This was designated as the "PAH-Soil" for the composting study. An equally large amount of arsenic-cyanide contaminated soil was also excavated and securely deposited. The major contaminants of these two soils are shown in Table 1, the result of a U.S. EPA Priority Pollutant Scan.

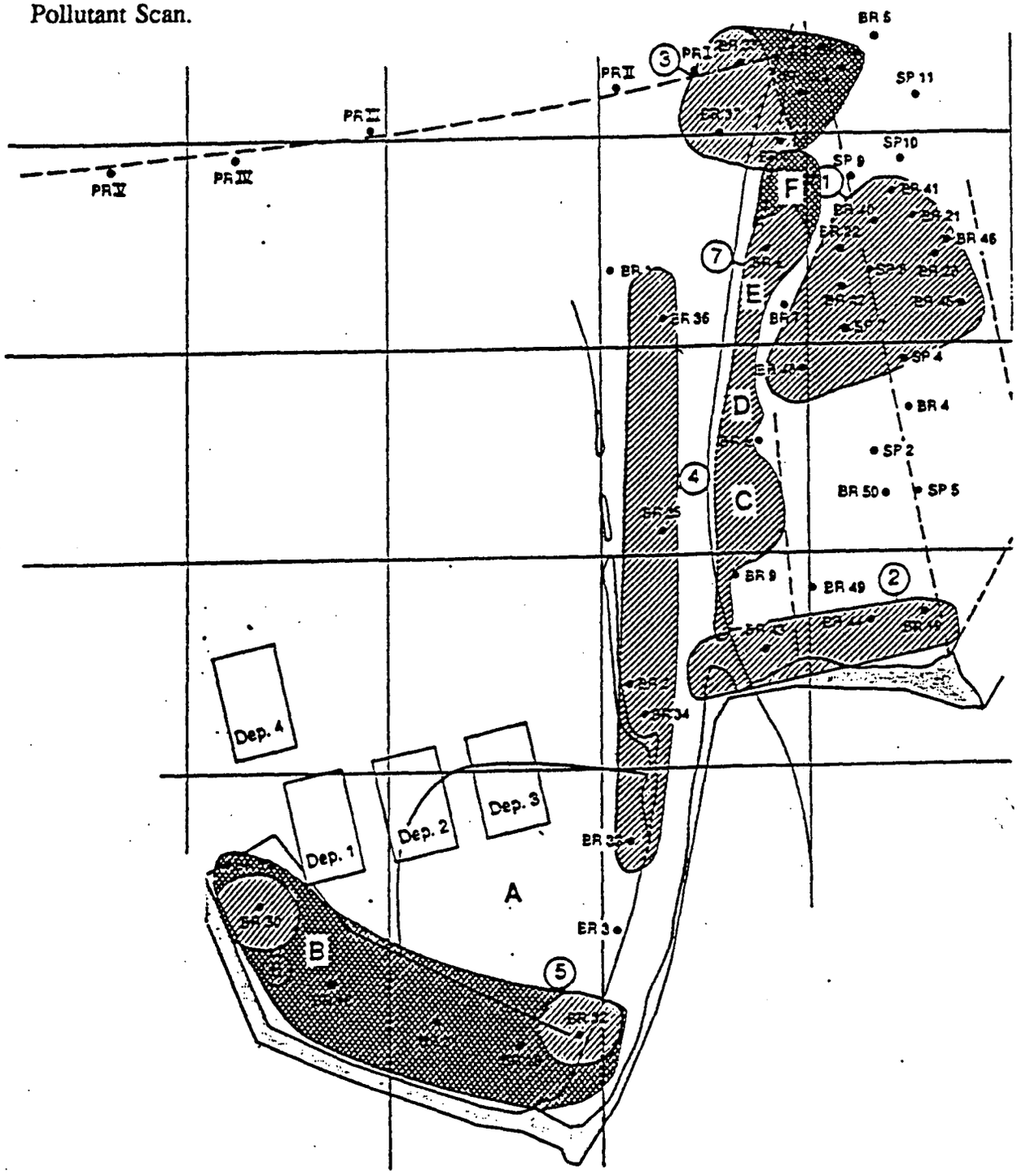


Figure 2. ***Site characterization during 1988-1990 revealed seven areas requiring remediation. Excavated soil was placed in secure deposits (Dep. 1-4) and comes mostly from two locations in Area No. 1, which still has significantly groundwater contamination.***

Table 1. **Major contaminants of the two excavated soil types and groundwater associated with the areas.**

	water 1	soil 1	water 2	soil 2
Compound	mg/l	mg/kg	mg/l	mg/kg
Arsenic	0.047	19.1	8.23	9970
Cyanide	0.136	2.3	2.45	1090
Naphthalene	11.1	381	0.004	0.667
3-methylnaphthalene				
2-methylnaphthalene	1.35	38.676		0.17
1-methylnaphthalene				
Biphenyl				
Sum bicyclic aromatics	12.45	419.676	0.004	0.837
Acenaphthylene	0.023		0.002	0.365
Acenaphthene	0.875	17.906	0.075	1.778
Fluorene	0.125	6.489	0.021	0.524
Phenathrene	0.021	2.288		1.986
Anthracene		0.211		0.441
Fluoranthene		0.283	0.002	1.77
Pyrene		0.187		1.748
Benzo (J,K) Fluoranthene				
Benzo (E) Pyrene				
Benzo (A) Pyrene				
Perylene				
Dibenzofuran	0.528	13.806	0.028	0.583
Sum PAH w/bicyclics	14.022	460.846	0.132	11.701
Sum PAH wo/bicyclics	1.572	41.17	0.128	9.195[1)]
Benzene			0.03	
Toluene	0.006		0.008	

More comprehensive testing of the PAH soil revealed that there were predominantly 17 PAH compounds and most of these were 2 and 3 ring structures (Table 2). The more complex PAH's were generally at concentrations which were below the proposed target treatment level, which was the Dutch "B" level of 20 mg/kg for total PAH's.

Table 2. Common PAH's at the Norsk Koksverk site.

Structure	Name	M.W.	Sol. (ug/l)	K_{ow}
	Naphthalene	128.19	30000	3.37
	2-Methylnaphthalene	142.20	----	----
	1-Methylnaphthalene	142.20	----	----
	Biphenyl	154.21	7500	3.95
	Acenaphthylene	152.21	3930	4.07
	Acenaphthene	154.21	3420	3.92
	Fluorene	166.23	800	----
	Phenanthrene	178.24	435	4.46
	Anthracene	178.24	59	4.5
	Fluoranthene	202.26	260	5.03
	Pyrene	202.26	133	4.98
	Benzo(j)fluoranthene	252.32	2.4	6.21
	Benzo(k)fluoranthene	252.32	2.4	6.21
	Benzo(e)pyrene	252.32	2.4	6.21
	Benzo(a)pyrene	252.32	3.8	6.04
	Perylene	252.32	2.4	6.21
	Dibenzofuran	168.2	10000	4.12

Source: Afghan and Chau. Analysis of Trace Organics in the Aquatic Environment, CRC Press 1989.

PILOT STUDY REMEDIATION PLAN

General

As stated above, there were several types of contamination, requiring different remediation processes. Three separate pilot treatability studies were conducted:

(1) Composting of excavated PAH-soil.
(2) Stabilization of excavated As-soil.

(3) Physical-Biological Pump-and-Treat of As-PAH groundwater.

The first study is reported herein.

Soil Composting Study

PAH-contaminated soil (Avg. concentration = 500 mg/kg total PAH) from "Dep. 4" was sorted, crushed, and mixed to form as homogenous a material as possible (Fig. 3).

The soil was then placed in 9 separate piles of ca. 10 m^3, 2 m x 3 m x 1.5 m (W x L x H), on geomembranes. Seven piles were placed in an unused industrial building, while two were placed in an abandoned local mine. The latter was chosen since the mine is a candidate full-scale treatment facility with excellent capacity for final, secure deposition of treated soil (Volume of storage space is 1 million m^3 with excellent ventilation and controlled drainage).

Table 3 shows the variables tested in the study, which are described below.

Table 3. Experimental variables in the pilot study. FA = Forced aeration, T = pile turning, N & P = Nitrogen and phosphorous.

Pile	Treatment				
	Bark	N&P	Aeration	Temp(°C)	Other
1.	-	-	T	10 - 16	-
2.	+	-	T	10 - 16	-
3.	+	+	T	10 - 16	-
4.	+	++	T	10 - 16	-
5.	+	-	FA	10 - 16	-
6.	+	+	H_2O_2	10 - 16	Recirc H_2O + dispersant
7.	+	+	T	25 - 35	-
8.	+	+	-	4	-
9.	+	+	T	4	-

Figure 3. *PAH-soil from "Dep. 4" (top panel) was sorted, crushed, and homogenized at the site (bottom).*

Bark Addition

Pine park was added to all piles except No. 1 (control with no amendments) in a ratio of bark: soil equal to 1:1 on a volume basis. The soil was sandy and had very little capacity to retain moisture.

Nutrients

Nitrogen and phosphorous were added to six of the piles in two different doses at the start of the study and after 8 weeks.

Oxygen

The piles were oxygenated by either turning the piles every three weeks, by forced aeration, or by peroxide addition via a water recirculation system. Peroxide was replenished three times per week.

Temperature

Ambient temperature ranged from 4-16°C for six of the piles in the industrial building. One pile was artificially heated by electric cables under the geomembrane base. The 2 remaining placed piles in the mine remained at a constant 4°C throughout the study.

Moisture

The piles were watered initially, and after weeks 6 and 8. Pile 6 also had regular periodic recirculation of water throughout the study. Dispersant, peroxide, and nutrients were added to the water.

Sampling and Analytical Methods

The piles were sampled twice weekly from 3 random locations at ca. 80 cm depth. Composite samples were prepared and placed in acid-washed brown glass jars and either analyzed immediately or frozen at -18°C. Temperature and soil gas measurements were taken at five locations at ca. 80 cm depth twice weekly also.

Analyses were conducted on site if possible. However, contract laboratories performed all PAH analyses. Analyses consisted of:

Moisture
pH

Tot N
Tot P
Total PAH (+ all components by GC/MS)
Soil gas (O_2 and CO_2)

RESULTS

PAH Treatment

The results of the study for the most predominant PAH's are shown in Figures 4-6. The group parameters, "Bicyclic aromatics", and "Total PAH" are shown in FIgures 4 and 5. In all cases, biphasic reduction in PAH's occurs over the 14 week study period. It is largely the duration of the lag phase or initial reduction rate that is influenced by the various amendments. Notably, the control pile shows the slowest rate of PAH reduction in all cases. The proposed treatment goal, the Dutch "B" level of 20 mg/kg PAH is achieved in 6-8 weeks under optimal conditions. The individual PAH's, as typified by Figure 6 for fluorene and acepnapthene, also follow the same behaviour. Forced aeration and nutrient additions both contributed to a much more effective process. Other lab experiments (data not shown) indicate that increased volatilization of the 2-5 ring PAH's by forced aeration was not significant, suggesting that it was primarily more effective biological activity that explains the reduction in PAH's.

Also, it is interesting to note that even at 4°C, there was effective removal of PAH's (See Fig. 6, Piles 8 and 9 for fluorene and acenapthene) suggesting that the naturally occuring populations had been well adapted to the low temperature environment. Owing to problems with regulating the temperature in the pile with heating cables, no reliable data were obtained for greater than ambient temperature which ranged from 4°-16°C during most of the study.

Lastly, the PAH removal results of the water recirculation experiment were unexpectedly low. Therefore, other dispersants were subsequently evaluated in bench scale batch and flow-through column studies. Results showed that another type of dispersant, ECO/+ (R.L. King Assoc. - Dutch Pride Products, 500 Airport Blvd., # 238, Burlingame, CA 94010) greatly enhanced the mobilizaton and removal of PAH's. In batch mixing studies, low concentrations of ECO/+ at ca. 10°C removed > 62 % of Total PAH's. The product is reported to be biodegradable so that the composting process should not be inhibited. Column studies are underway to test washing and biodegradation effects simultaneously.

The results of the PAH reduction aspects of the study are compared with available published literature values in Table 4. Generally, the results from the Koksverk pilot study are comparable with the published studies. Where it is possible to directly compare individual compounds, for example with phenanthrene, the half life (t ½) in this study was ca. 14 days under optimal conditions versus 16-200 days under a range of other comparable conditions of temperature (10-20°C) and amendments (added nutrients). The same is true for fluoranthrene, with this study reporting t ½ = 48.5 days versus 29 to 440 days. (Sims, et al, 1988; Sims, 1986; and Coover and Sims, 1987).

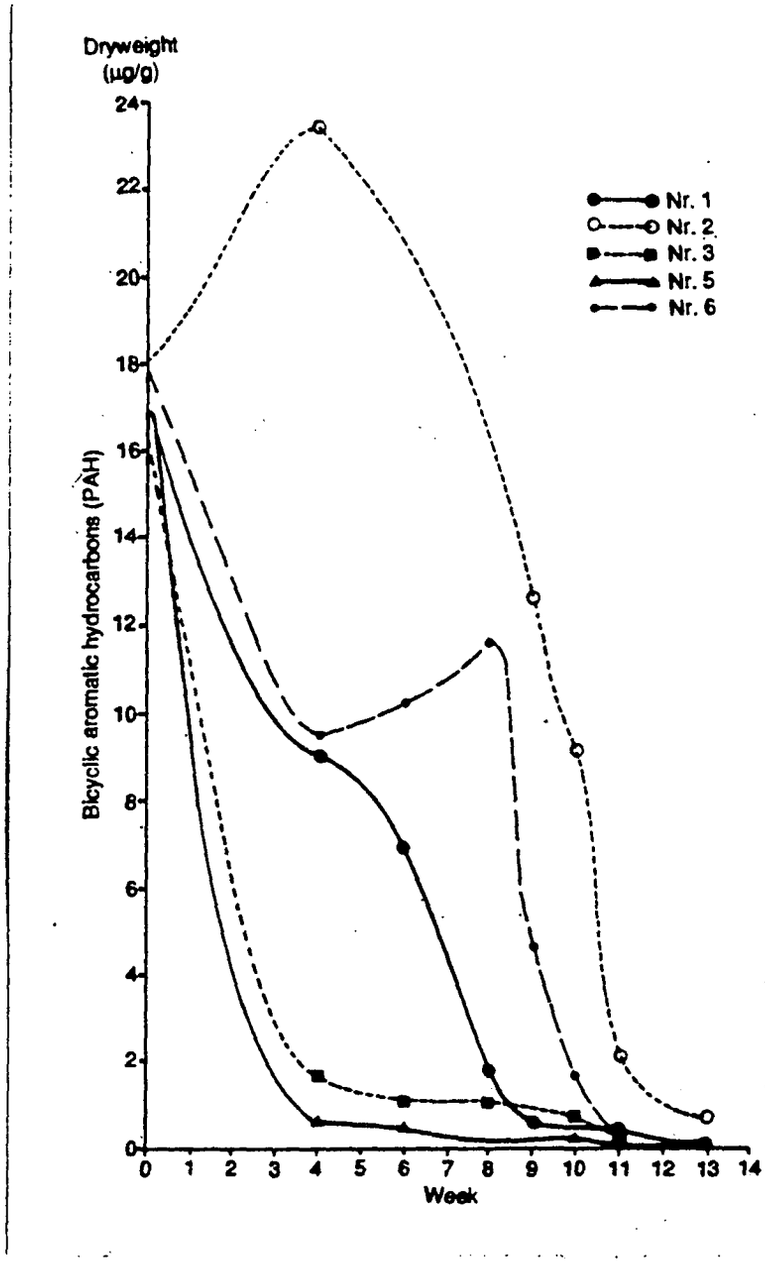


Figure 4. Reduction in bicyclic aromatics.

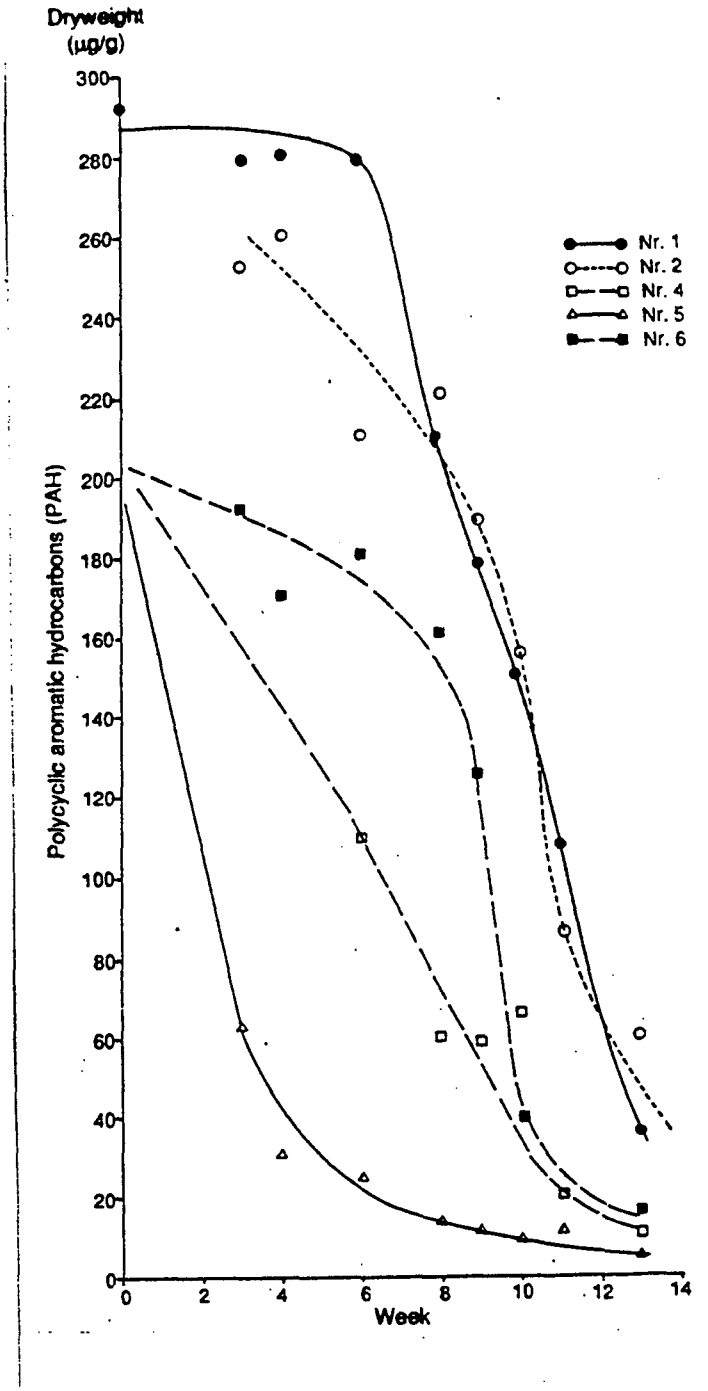


Figure 5. Reduction of Total PAH's.

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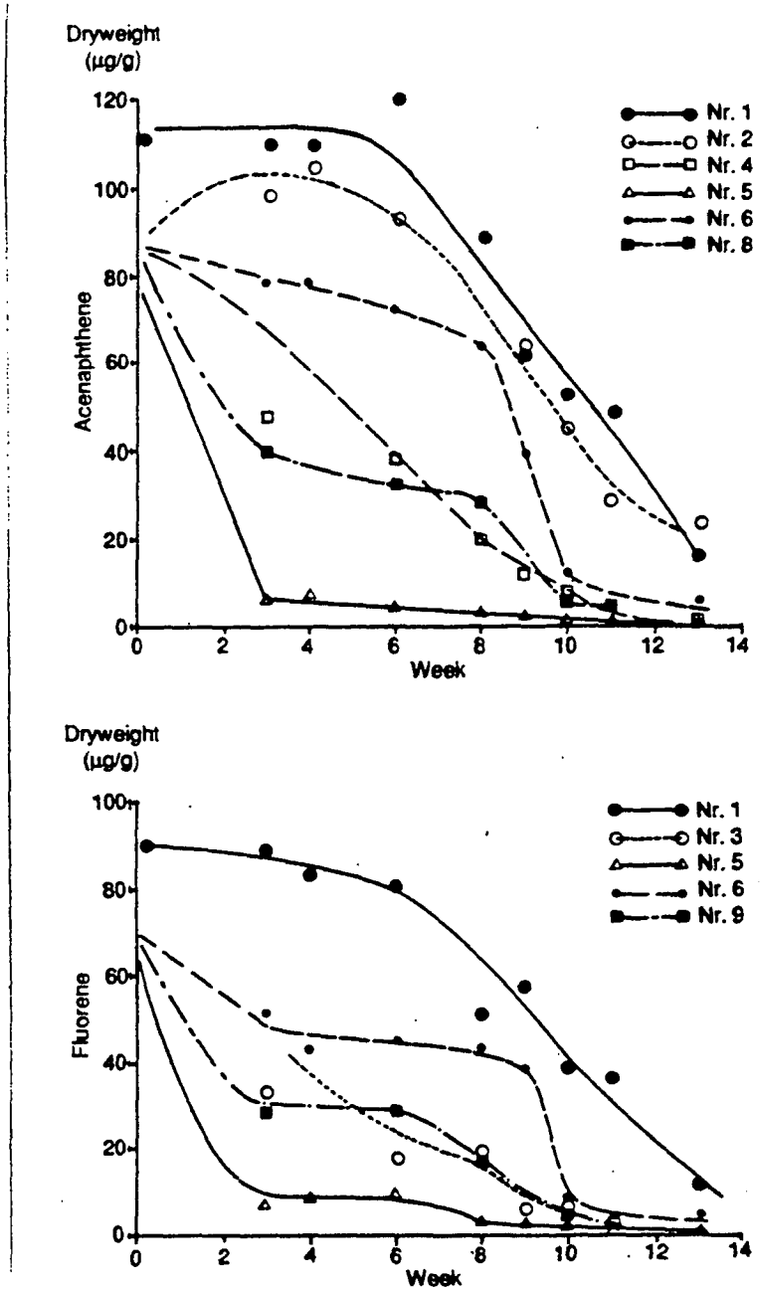

Figure 6. Reduction of fluorene and acenaphthene.

Where total PAH data were available, this study reports t ½ = 22 days under optimal field conditions versus 43 days in a laboratory study (McGinnis, et al, 1991).

Table 4. Results of this study (Norsk koksverk, 1990) are compared with literature values for half-lives (t ½) from similar laboratory experiments or field studies.

Half-lives of Selected PAH's.

Group/Compound	Conditions	C_0 (mg/kg)	$t_{½}$ (days)	Source
Naphthalene	20°C Resp. Test	101	2.1	Sims et al., 1988
1-Methylnaphthalene	20°C Resp. Test	102	1.7	Sims et al., 1988
Σ Bicyclics	4-16°C Pilot scale + amendments	18	19.2	Norsk koksverk, 1990
	4-16°C Pilot scale No amendments	24	87.7	Norsk koksverk, 1990
Phenanthrene	20°C Resp. Test	902	16	Sims et al., 1988
	Field Soil - No amend.	—	69	Sims, 1986
	Field Soil + amend.	—	23	Sims, 1986
	10°C Resp. Test	—	200	Coover and Sims, 1987
	20°C Resp. Test	—	60	
	4-16°C Pilot scale + amend.	40	13.9	Norsk koksverk, 1990
	4-16°C Pilot scale - No amend.	54	90.1	Norsk koksverk, 1990
Flouranthene	20°C Resp. Test	883	377	Sims et al., 1988
	Field Soil - No amend.	—	104	Sims, 1986
	Field Soil + amend.	—	29	Sims, 1986
	20°C Resp. Test	—	440	Coover and Sims, 1987
	4-16°C Pilot scale + amend.	4	48.5	Norsk koksverk, 1990
Σ PAH	22° EPA Lab. procedure	1095	43	McGinnes et al., 1991
	4-16°C Pilot scale + amend.	260	22.2	Norsk koksverk, 1990
	4-16°C Pilot scale - No amend.	290	98.4	Norsk koksverk, 1990

OPERATING PARAMETERS

1. Temperature

The development of pile temperatures in the building is shown in Figure 7. Pile 1 temperatures reflect the ambient air temperature which ranged from 0°C to 16°C at the end of the study. All piles showed greater than ambient temperatures due to biological activity, the highest being Pile No. 4 which had a very high dose of N & P. The temperature of the piles in the mine remained at 4°C throughout the study.

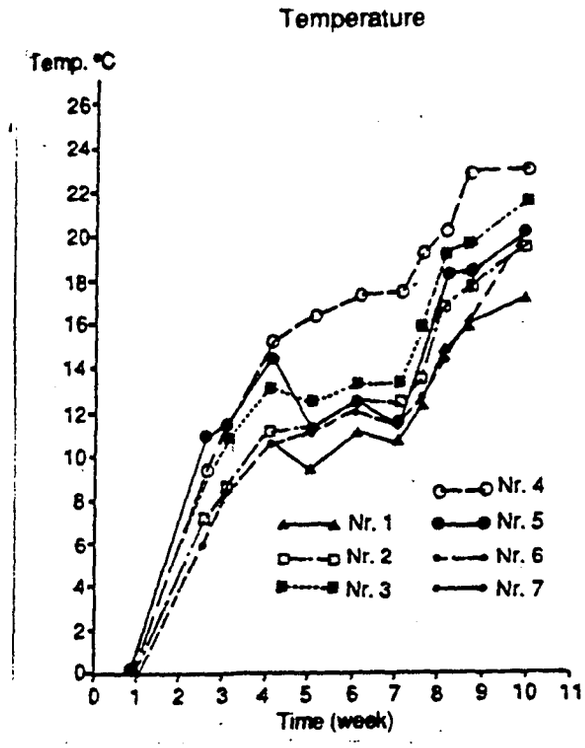

Figure 7. Temperature development in the compost piles.

2. Moisture

The percent moisture reported as an average of all samples are shown in Figure 8. Pile No. 1, the control, was ca. 7 % throughout the study. The other piles which had the added bark matrix ranged from 25 to 35 %. Pile No. 6 which received regular sprinkling with recirculated water retained ca. 32 % moisture after the watering program was started, which is considered the maximum attainable for this soil/matrix combination.

3. Soil gas O_2 and CO_2

The average soil gas values are also shown in Figure 8. The relationship between the O_2 and CO_2 values indicates the biological activity, as evidenced by PAH reduction, quite well. The control pile No. 1, for example, which exhibited the least biodegradation of PAH's shows the highest ratio of O_2: CO_2, whereas in the other piles the ratio is either close to one (Pile No. 2), or the relationship is reversed. The only exception is for Pile No. 5 receiving forced aeration, in which gas phase CO_2 is rapidly exchanged with O_2, hence yielding a low value.

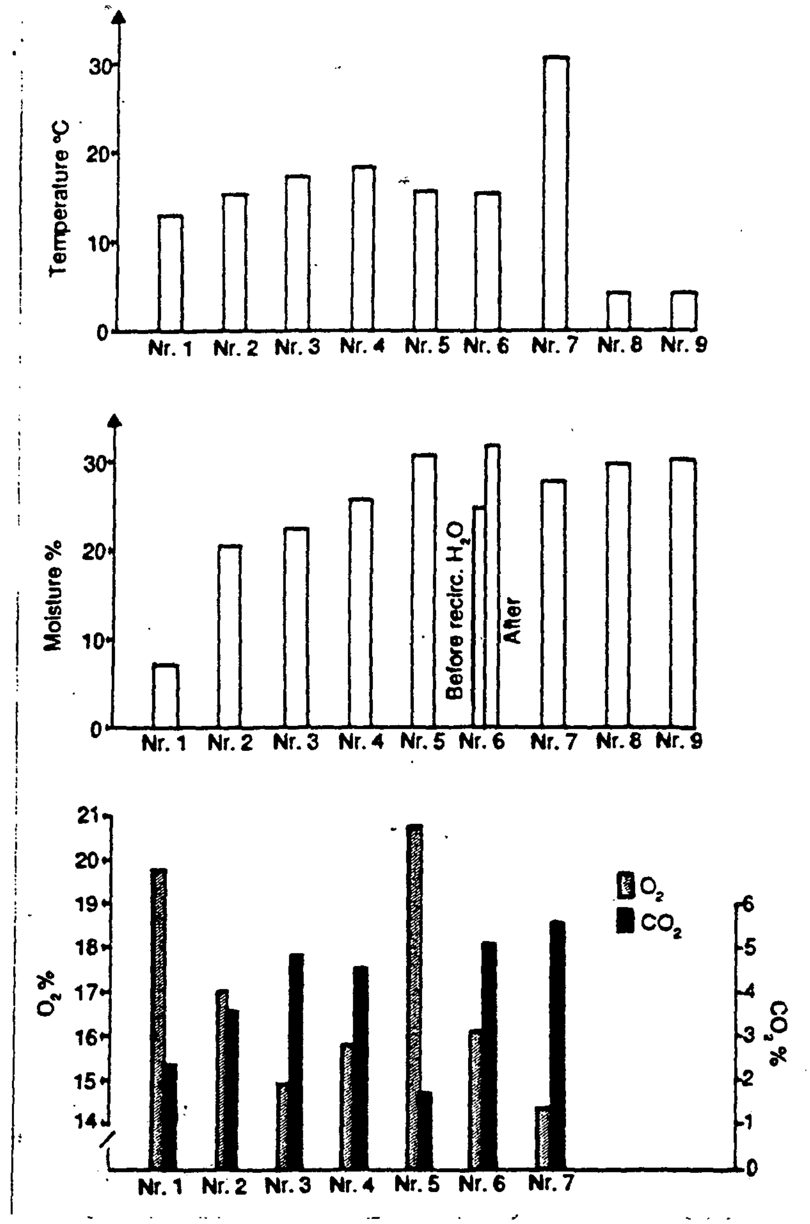

Figure 8. ***Average temperature (°C), moisture (%), and soil gas O_2 and CO_2 (%).***

4. Toxicity

Limited plant toxicity tests were conducted with rye grass which quantitatively showed that treated soil under optimal conditions was ca. 80% less inhibitory to plant growth relative to the untreated soil. This study was conducted since one alternative for ultimate disposal of the soil was as cover material at a local landfill.

Toxicity of groundwater from the same area from which the soil originated was also treated biologically in a parallel study. Toxicity was measured by standard "Microtox™" procedures

and showed a 93 % reduction in toxicity of the treated water as compared to the raw groundwater (data not reported here). It is assumed that leachate from the soil composting studies will behave similarly, thus some form of toxicity testing is recommended as a routine quality control parameter during full-scale operations.

SUMMARY

Pilot Study Results

1. Among the amendments evaluated in the study, and addition of bark and nutrients, primarily nitrogen and forced aeration, essential for optimal biological activity.

2. The surfactants chosen for the pilot study did not improve PAH removal. However, subsequent column and batch studies with another commercially available product (ECO/+) were very promising.

3. Cultures adapted to low temperatures showed significant degradation at 4°C, however, better results were obtained at temperatures ranging from 6-16°C, as one would expect.

4. The proposed treatment objective of 20 mg/kg Total PAH was attained within 6-7 weeks, while a more stringent goal of 10 mg/kg was reached within 8-9 weeks.

Considerations for Full-scale Remediation

1. **Location.** The site is large enough to accomodate composting remediation on site for the 20,000 tons of excavated soil. However, if development of the site is to proceed quickly an attractive alternative is an abandoned mine. It is scheduled to be a full-scale treatment facility with a 1 million m^3 capacity for secure deposition of wastes.

2. **Facilities.** On site composting will require tents placed over geomembrane liners. Operations in the mine will require only the impervious liner.

3. **Operations.** After sorting and crushing, samples will be taken to ensure that the arsenic contamination is below acceptable limits. High As-containing soil will be separately treated by stabilization. Then the soil will be mixed with bark and nutrients and dispersant, and placed in windrows at the treatment facility.

   Forced aeration will be used if site development plans require a 30-40 % shorter treatment program. Capacity for air heating will be designed in the system. Aeration will not commence until the third or fourth week of operation, when most of the semivolatile bicyclic aromatics have been degraded.

ACKNOWLEDGEMENTS

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