

## **Air pollution of hydrocarbons exhausted from vehicle in tunnels, bridges of Istanbul and detection of 3-nitrophthalic in seawater near the side of boat**

### **Istanbul'da tunnel ve köprülerde taşıt egzozundan çıkan hidrokarbonların sebep olduğu hava kirliliği ve deniz motoru yanındaki suda 3-nitrofitalik asit tayini**

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#### **Abstract**

The air pollution due to exhausted gas from vehicles was studied in tunnels, bridges in Istanbul city and the contribution of exhausted gas from boat to the seawater in Golden Horn. The component of hydrocarbons of fuels and exhausted gas from vehicles was also compared. The gas samples were collected in tubes containing Tenax GR or activated charcoal adsorbent. They were extracted with dichloromethane and analysis performed by UVF, GC/MS and HPLC. The petroleum hydrocarbon amounts in gas exhausted were from diesel car II 89.40  $\mu\text{g}/\text{m}^3$ , from car I 13.43  $\mu\text{g}/\text{m}^3$ , from normal gasoline car 41.43  $\mu\text{g}/\text{m}^3$  and from super gasoline car 32.10  $\mu\text{g}/\text{m}^3$ . The highest hydrocarbon pollution was found in Haşim İşcan tunnel and hydrocarbon amount ranged as 5.30-11.00  $\mu\text{g}/\text{m}^3$ . During the analysis over 22 aliphatic, 23 aromatic hydrocarbon and nicotine were identified in Haşim İşcan tunnel. In this area carcinogenic, aromatic hydrocarbons, benzene derivatives and toluene were detected. The highest hydrocarbon pollution was found on Atatürk and Galata bridges as 9.65  $\mu\text{g}/\text{m}^3$  and 8.36  $\mu\text{g}/\text{m}^3$  respectively.

For the first time a new nitro-derivate, 3-nitrophthalic acid was detected in seawater near the exhaust pipe of boat and it was proved by GC/MS and HPLC analyses.

The adsorbent capacity of Tenax GR with charcoal compared and Tenax GR adsorbed better than charcoal.

**Keywords:** Istanbul city, tunnels, bridges, hydrocarbon pollution, seawater, 3-nitrophthalic acid.

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## Introduction

The increase of air pollution is a main problem for environment. Among the pollutants, petroleum hydrocarbons are important due to their high toxicity and carcinogenicity. The origin of petroleum hydrocarbons in air are combustion of natural gas, coals, fuels oil, forest, prairies fires and exhausted gas from vehicles. The hydrocarbons are released into atmosphere as burned/changed and not burned/unchanged forms.

Numerous reports have appeared on the air pollution in the various cities of the world as Mexico City (Salazar *et al.*, 1991), Nagasaki (Japan) (Wada *et al.*, 2001), Dublin, Ireland (Broderick and Marnane, 2002), La plata (Argentina) and Leipzig (Germany)(Rehwagen *et al.*, 2004), Hangzhou (China) (Zhu *et al.*, 2004), Taiwan (Yang and Chen, 2004), Brisbane (Australia) (Mc Kenzie *et al.*, 2005; Lim *et al.*, 2005), Seoul (Lee *et al.*, 2005), Athens (Greece) (Chatzis *et al.*, 2005), Hong Kong (Xia and Shao, 2005), Chicago (Li *et al.*, 2005), Sao Paulo (Bourotte *et al.*, 2005).

The ships have also a role for air and marine pollution (Ijlstra, 1990). There are a few literatures on the contribution of air pollution to sea water (Hoffman *et al.*, 1984).

The other problem in the air pollution is nitrogen oxide (NO)<sub>x</sub>. It is generally produced by conversion of nitro monoxide which originates from care (Lee, 2005) and also the ships (Ijlstra, 1990). The main chemical reaction of nitrogen oxide with hydrocarbon radicals gave nitro-compounds. The determined nitro PAHs compounds were: 1-nitropyrene (Samanta *et al.*, 2002), 1, 3- dinitropyrene, 1, 6- and 1, 8- dinitro pyrene. Nitro- and dinitro/PAH are found in diesel exhaust (Pohjola, 2004). Nitro-PAH is carcinogenic/ mutogenic compounds (Hansen *et al.*, 2004). The other carcinogen compounds are benzene, 1,3-butadiene (Broderick and Marnane, 2002), benzo[a]pyrene, dibenzo[a,h]anthracene, dibenzo[a,l]pyrene (Okana-Mensah *et al.*, 2005).

Istanbul city is situated on the two side of the narrow channel of Bosphorus which connects Europe and Asia. Istanbul city has an area of 5512 km<sup>2</sup> with an approximate population of 13 million habitants and in 2005 the total vehicles number are 2212310.

Daily traffic flow in the studied area at the Haşim İşcan tunnel is between 07-09 hours are: 400 car, 45 bus in 10 min. through Taksim direction and 360 car, 46 bus in 10 min. through Aksaray direction. 12 boats flow between Eminönü-Karaköy visa versa in Golden Horn.

In this work, we investigated exhausted gas from vehicles and their influence on air pollution in tunnels and bridges in Istanbul city and the contribution of exhausted gas from boats in Golden Horn seawater.

### **Material and Method**

The air samples were taken in tunnels, bridges which are a major site of Istanbul and also in seawater near the side of boats at Golden Horn. The sampling sites are shown in Figure 1. No industrial plants are located in examined area.

Sampling places and dates are;

1. Tunnels: 1.1. Haşim İşcan 06 and 11.04.2005, 1.2. Edirnekapı 08.06.2005
2. Yenikapı bus platform 27.04.2005
3. Bridges in Golden Horn: 3.1.Haliç Bridge 08.06.2005, 3.2.Atatürk Bridge 10.06.2005, 3.3.Galata Bridge 10.06.2005
4. Boats: Seawater sample near the side of boats at Galata Bridge, (Eminönü, Golden Horn) 18 and 25 April 2005.
5. Vehicles: 12.05.2005

Air samples in tunnels were taken in morning at peak hours of traffic.

The samples were collected with an air sampler (Gilian LFS-113, USA) connected to a stainless steel tube with a diameter of 0.5 cm, a length of 10 cm. Adsorbents used in tubes are Tenax GR 60/80 or activated charcoal. Sampling was conducted at a flow rate of 350 cm<sup>3</sup>/min. The collected sample volume is 3500 cc in 10 min.

Exhausted gas from gasoline/diesel vehicles (car and bus) were collected by a glass funnel positioned at the exhaust pipe connected to adsorbent tube. After taking the air samples, the tube is immediately capped with cover and transported to the laboratory. The elution was made three times with 20 ml of dichloromethane (DCM). The extracts were combined then distilled at 36°C. The residue was taken with hexane and hydrocarbons amount determined by UVF and its components were analyzed by GC/MS. 3-nitrophthalic acid was analyzed by GC/MS and HPLC.

3 L Seawater sample was taken at surface water of 20 cm near the boats exhaust pipe.

The gasoline and diesel fuel were obtained from the station for plotting of calibration curves and also the comparison their hydrocarbon component with exhausted gas from vehicles.

Figure 1. Sampling stations

## *Analyses*

### *1-UVF analysis for hydrocarbon pollution in air*

The calibration curve of normal/super gasoline and diesel fuel were plotted in a concentration of 0.25-2 µg/ml and 0.5-2 µg/ml in hexane respectively by UVF (Shimadzu, 1601) at 310/360 nm (ex/em) and its equation was taken from apparatus. The air pollution amount in examined samples was calculated through each equations of tested fuel equivalent.

### *2-GC/MS analysis*

GC/MS analysis: GC (HP 6890) coupled to mass spectrophotometer HP 5972 A. A split/splitless injector was used, injection; 2 µl, split time: 1 min<sup>-1</sup>, flow 37 ml min<sup>-1</sup>. Column; HP-5MS: 30.5 m x 250 µm x 0.25 µm nominal. The injector temperature was maintained at 280°C. The GC temperature programme was: from 40°C to 280 °C at 8 °C min<sup>-1</sup>. The carrier gas was helium, flow rate 1 ml min<sup>-1</sup>.

### *3-HPLC analysis*

HPLC (HP 1100) UV-DAD detection was used. The chromatographic column was C<sub>18</sub> (Waters) 3.9 x 150 mm. Elution was carried out at a flow rate of 1 ml min<sup>-1</sup> using a gradient of mobile phase acetonitrile-water starting, 5 min from 40-60 (V/V) to 100 % acetonitrile over 25 min.

Chemicals: Reference compound of 3-nitroptalic acid was purchased from Acros Organic (Belgium). Tenax GR 60/80 (Gilian LFS-113, USA), Activated charcoal (Norit, Holland).

Solvent: Dichloromethane (HPLC grade Lab Scan, Ireland). Hexane (Merck, Germany).

## **Results and Discussion**

The quantity of air sample examined in the literature is ranged 1 to 10 L. We examined 3 L. The petroleum hydrocarbon amount was calculated in air as µg/m<sup>3</sup>.

### *1- Results of fuel and exhausted gas from vehicle*

The petroleum hydrocarbon amounts determined by UVF in exhausted gas from vehicles are shown in Table 1.

**Table 1.** The petroleum hydrocarbon amount of exhausted gas from vehicles ( $\mu\text{g}/\text{m}^3$ ).

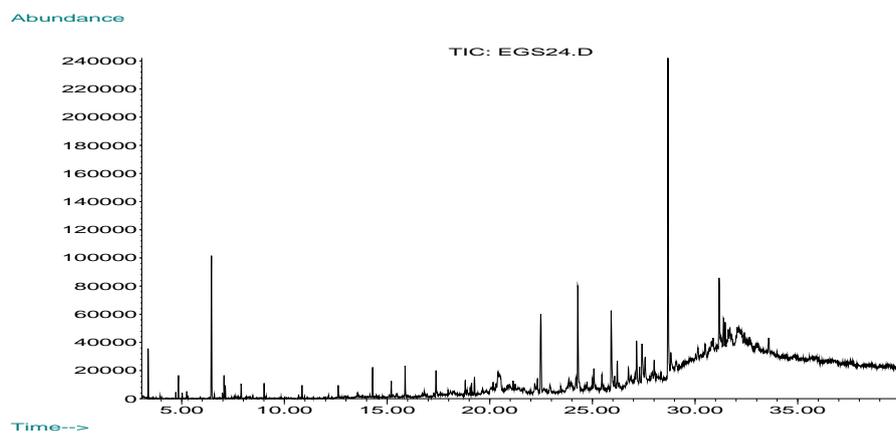
Fuel and vehicles types	Petroleum hydrocarbon amount found in exhausted gas
Normal gasoline car	<b>41.43</b>
Super gasoline car	32.10
Diesel car 1	13.43
2	<b>89.40</b>
Bus 1	21.95
2	12.69
3	11.46
Boat (Diesel)	25.94

As can be seen in Table 1, emission profile of petroleum hydrocarbon can be affected by chemical composition of fuel and also motor types. A large difference on hydrocarbon amount were observed between diesel and gasoline exhaust.

According to these findings the highest gas emission is ranked as diesel vehicle 2 followed by normal gasoline car.

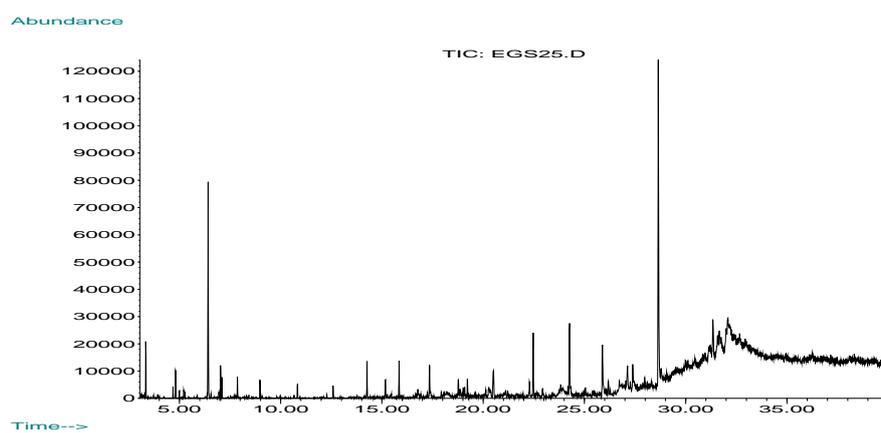
The chromatogram of vehicle fuel and exhausted gas are shown in Figure 2-8.

**Figure 2.** The chromatogram of diesel fuel I.



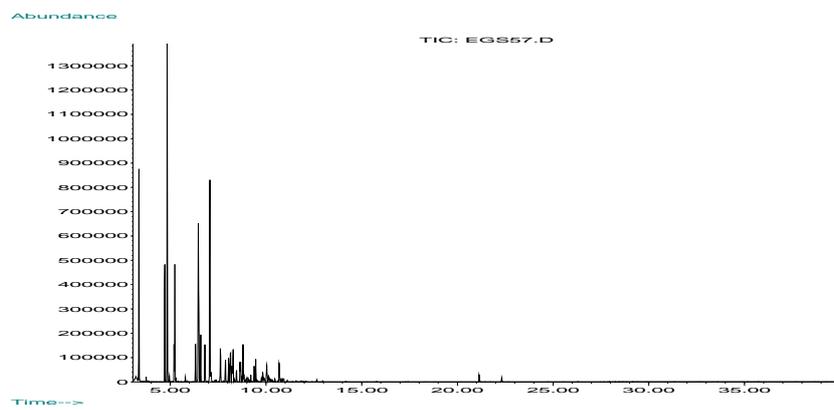
**Figure 3.** The chromatogram of exhausted gas from car fuelled diesel I.

**Figure 4.** The chromatogram of diesel fuel II.



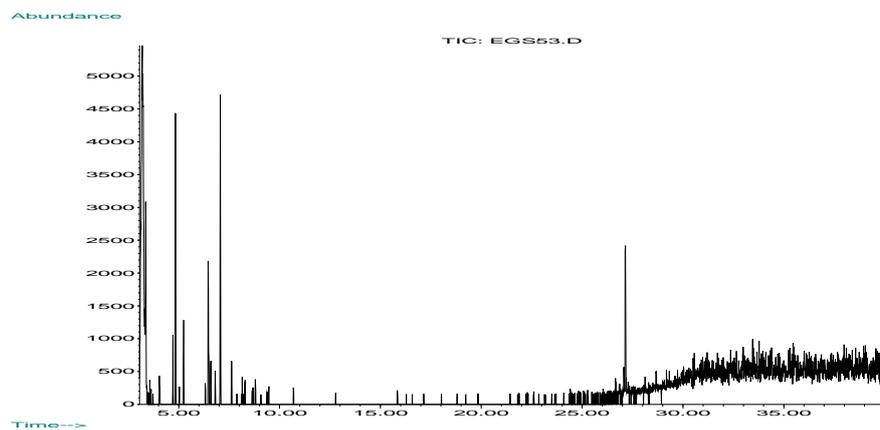
**Figure 5.** The chromatogram of exhausted gas from car fuelled diesel II.

**Figure 6.** The chromatogram of normal gasoline.



**Figure 7.** The chromatogram of exhausted gas from car fuelled normal gasoline.

**Figure 8.** The chromatogram of super gasoline.



**Figure 9.** The chromatogram of exhausted gas from car fuelled super gasoline.

The hydrocarbon composition of exhausted gas from diesel car I:

Decane	Benzaldehyde
Dodecane	Ethylbenzene
n-Tetradecane	Toluene
Pentadecane	m-Xylene
Heptadecane	o-Xylene
Hexadecane	p-Xylene
Hexadecanoic acid	Phenol
Undecane	Butylphthalate
Cycloheptatriene	Ethylphthalate
2,4,6-Trimethyloctane	

The hydrocarbon composition of exhausted gas from diesel car II:

7-Methyl-3-octen-2-one	Docosane
1-Nonanol	Toluene
Decane	o-Ethyltoluene
Dodecane	1-Ethyl-2,4-dimethylbenzene
Tridecane	Ethylbenzene
2-Tridecanol	isopropylbenzene
Heptadecane	m-Xylene
9-Octadecane	Phenol
Nonadecane	DEHP
Undecane	n-Butylisobutylphthalate

The hydrocarbon composition of exhausted gas from normal gasoline car II:

Cyclooctatetraen	Phenol
Octane	Toluene
Nonane	2-Propyltoluene
Undecane	o-Ethyltoluene
2-Methylpropenylbenzene	m-Ethyltoluene
1,2,3-Trimethylbenzene	m-Propyltoluene
1,2,4-Trimethylbenzene	m-Xylene
1,3-Dimethyl-2-ethylbenzene	3-Ethyl-o-xylene
1-Isopropyl-3,5-dimethylbenzene	4-Ethyl-o-xylene
1-Isopropyl-4-dimethylbenzene	2,5-Dimethylstyrene
1-Methyl-2-propylbenzene	1-Methylindane
n-Butylbenzene	1,3-Dimethylindane
n-Propylbenzene	1,6-Dimethylindane
Isobutylbenzene	4,6-Dimethylindane
Isopropylbenzene	1H-Indene
m-Diethylbenzene	Napthalene
Sec-butylbenzene	2-Methylnapthalene
C3 benzene	Isobutylphthalate

The hydrocarbon composition of exhausted gas from fuelled super gasoline car:

Decane	Sec-butylbenzene
2,6,10-Trimethyldodecane	Toluene
3,6-Dimethylundecane	m-Ethyltoluene
Pentadecane	o-Ethyltoluene
Hexadecane	p-Ethyltoluene
Heptadecane	n-Propylbenzene
Nanodecane	p-Diethylbenzene
Hexadecanoic acid	m-Xylene
Eicosane	p-Xylene
Benzene	3-Ethyl-o-xylene
C3-benzene	Isooctylphthalate
Phenol	
1,2,4-Trimethylbenzene	
1,3,5-Trimethylbenzene	

Fatty acids as hexadecanoic, octadecanoic and linoleic acid were detected in exhausted gas of Yeni Kapı bus station is due to biodiesel fuel.

## 2- Results of tunnels

The air pollution in tunnels is shown in Table 2. The hydrocarbon pollutions were calculated through different fuel equivalent examined.

**Table 2.** Petroleum hydrocarbon pollution calculated through different fuel equivalent (Eq) in tunnels  $\mu\text{g}/\text{m}^3$ .

Collection Date	Diesel Eq.	Normal gasoline Eq.	Super gasoline Eq.	Mean values of diesel and gasolines Eq.
Haşim İşcan (06/04/2005)(a)	5.82	5.98	7.60	6.29
Haşim İşcan (06/04/2005)(b)	5.78	6.57	8.30	6.88
Haşim İşcan (11/05/2005)(a)	7.97	8.86	10.88	9.23
Haşim İşcan (11/05/2005)(b)	8.46	9.79	11.10	9.78
Edirne Kapı (08/06/2005)	6.19	7.07	8.75	7.33

(a) Through Taksim direction, (b) Through Aksaray direction

The amount of hydrocarbons in Haşim İşcan tunnel were varied depending on fuel equivalent as 5.30-8.46  $\mu\text{g}/\text{m}^3$  through diesel and 7.60-11.10  $\mu\text{g}/\text{m}^3$  through super gasoline, 5.98-9.79  $\mu\text{g}/\text{m}^3$  through normal gasoline equivalent.

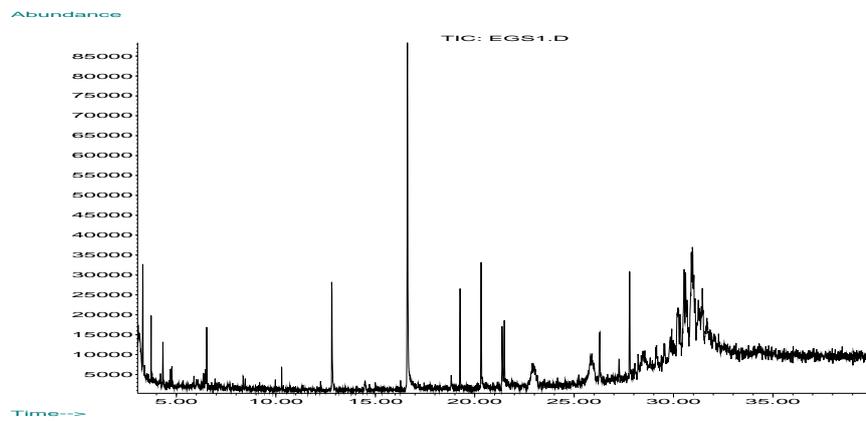
The highly dense traffic in Haşim İşcan tunnel were responsible for the extreme value of pollution.

In Edirnekapı Tunnel the traffic was not high also the oil pollution was low.

As seen in Table 2 the oil levels in air through the calculation from diesel equivalent are low while the number of car is low.

On the other hand nicotine was detected in Haşim İşcan tunnel which are many smokers at this area.

GC/MS chromatogram of Haşim İşcan tunnel is shown in Figures 10.



**Figure 10.** GC/MS chromatogram of air sample taken in Haşim İşcan tunnel.

Hydrocarbon components detected in air of Haşim İşcan tunnel taken in various dates are:

2,3-dimethyl-2-pentene	1,2,4-trimethylbenzene
2,2,3,4-tetramethylpentane	1,3,5-trimethylbenzene
3-methyl-1-pentene	1-methyl-2-propylbenzene
1-octanol	Ethylbenzene
Nonane	Isopropylbenzene
4-methyl-5-propylnonane	Phenol
3,7-dimethylnonane	Benzaldehyd
Octane	Toluene
3-methylnonane	methyltoluene
4-hydroxy-4-methyl-2-pentanon	m-ethyltoluene
Decane	o-ethyltoluene
2-methyldecane	4-isopropyltoluene
4-methyldecane	o-xylene
5-methyldecane	m-xylene
5-methylundecane	p-xylene
Dodecane	3-ethyl-o-xylene
Butylcyclohexane	Naphthalene
2,2-dietoxypropan	Phenanthrene
1-heptadecanol	Butylphthalate
Undecane	Ethylphthalate
1,4-dimethylcyclohexane	Isooctylphthalate
1,2-dimethylcyclohexane	Diethylphthalate
C3 benzene	DEHP
Hexadecanoic acid	<i>Nicotine</i>

The hydrocarbons determined in air sample of Haşim İşcan tunnel taken in activated charcoal adsorbent:

Octane  
1-(2-butoxyetoxy)-ethanol  
1,2-dimethylcyclohexane  
1,4-dimethylcyclohexane  
2,4-dimethylhexane  
Ethylbenzene  
Hexandioic acid, bis(2-ethylhexil) ester  
o-xylene  
p-xylene  
1-formil-4-methylnapthalene  
Butylphthalate  
Diethylphthalate  
Dioctylphthalate  
Isobutylphthalate  
Isooctylphthalate  
N-butylisobutylphthalate

### 3- Results of bridges

Petroleum hydrocarbon pollution of air on bridges calculated from different oil equivalent are shown in Table 3.

**Table 3.** Petroleum hydrocarbon amount on bridges calculated from trough different equivalent (Eq)( $\mu\text{g}/\text{m}^3$ ).

Stations	Diesel equivalent	Normal gasoline equivalent	Super gasoline equivalent	Mean Values
Haliç Bridge (08/06/2005)	5.28	6.62	8.36	6.75
Atatürk Bridge (08/06/2005)	6.73	7.71	9.65	8.03
Atatürk Bridge (10/06/2005)	5.12	6.43	8.09	6.55
Galata Bridge (10/06/2005)	6.45	7.76	9.84	8.02

As can be seen in the Table 3 the highest air pollution is observed in Galata and Atatürk Bridge. The amount of hydrocarbons are changed depending sampling date.

### 4- Results of seawater

The hydrocarbon components of diesel boat and the contribution of exhausted gas from the boats to seawater are as follows.

Seawater taken from bow of the boat

5-Ethyl-2-methylheptane

2,3-Dimethylheptane

1-Octanol

Undecane

Tridecane

Tetradecane

3,8-Dimethyldecane

Pentadecane

Hexadecane

Heptadecane

Octadecane

Nonadecane

2,6,10,14-Tetramethyl-pentadecane

2,6,10-Trimethyldodecane

Eicosane

Heneicosane

Pentacosane

Dotriacontane

Tritriacortane

p-Xylene

1,3-Dimethylnapthalene

Seawater taken from 20 cm near the side of exhaust pipe of boat

Butyl-cyclohexane	Docasone
4-Methyl-3-penten-2-one	Heptacosane
Octane	Hexadecanoic acid
2,3,4-Trimethylhexane	9,12-Octadecadienoic acid
2,3,3-Trimethyloctane	Hexatriacontane
2,6-Dimethyloctane	Sec-butylbenzene
Nonane	1,2,3-Trimethylbenzene
Decane	1,3,5-Trimethylbenzene
Undecane	1-Methyl-2-isopropylbenzene
Dodecane	1-Methyl-4-(1-methylethyl)-benzene
1-Undecanol	Ethylbenzene
1-Dodecanol	Isopropylbenzene
2-Methyl-2-hexanol	Phenol
4-Methyldecane	m-propyltoluene
Tridecane	2-Propyltoluene
2-Tridecanol	o-Ethyltoluene
Hexadecane	o-Xylene
1-Hexadecanol	m-Xylene
2,6,10,14-Tetramethylhexadecane	p-Xylene
2,6,10,14-Tetramethylpentadecane	3-Ethyl-o-xylene
Tetradecane	4-Ethyl-o-xylene
Pentadecane	1H-indene,2,3-dihydro-1-methyl
Heptadecane	Di-(2-ethylhexyl)phthalate
Octadecane	Butylphthalate
Nor-pristane	n-Butylisobutylphthalate
Nonadecane	<i>3-Nitrophythalic acid</i>
n-Eicosane	
Heneicosane	

GC/MS chromatogram of 3-Nitrophythalic acid and its spectrum and their spectrum taken from HP memory are shown in Figure 11 and 12 respectively.

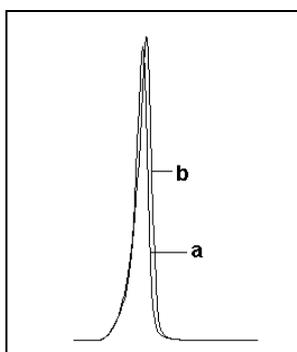
**Figure 11.** GC/MS chromatogram of 3-nitrophythalic acid from the near side of exhausted gas from the boats to seawater

**Figure 12.** Spectrums of 3-nitrophythalic acid (a) from the near side of exhausted gas from the boats to seawater and (b) taken from the HP memory

Mass spectra of 3-nitrophythalic acid  $m/z$  211, 167 ( $-\text{CO}_2$ ), 149 ( $-\text{H}_2\text{O}$ ), 103 ( $-\text{NO}_2$ ), 102 ( $-\text{H}^*$ ).

The 3-nitrophthalic acid was identified by GC/MS and the findings also confirmed by HPLC analysis. Figure 13 shows the peaks of superimposed the reference compound 3-nitrophthalic acid with the sample of seawater taken near the side of boat.

The important point in the present work is the detection of 3-nitrophthalic acid for the first time from seawater taken from near the exhaust pipe of diesel boat. The origin of 3-nitrophthalic acid is due to the reaction of  $\text{NO}_x$  with phthalic acid which exists in fuel as form phthalate ester.



a – reference compound

b – sample

**Figure 13.** The peaks superimposed on HPLC chromatogram of 3-nitrophthalic acid (a) formed in boats exhausted gas and reference compound (Acros) (b).

#### *5- Comparison of the adsorbent capacities*

The adsorption capacities of Tenax GR and of activated charcoal are also investigated and the results are shown in air of Haşim İşcan tunnel analysis.

As seen in these findings Tenax GR is more adsorbed aliphatic alkane, alkene, branched alkene and aromatic hydrocarbons than activated charcoal.

#### **Conclusion**

This is the first time, the determination of composition and concentration of hydrocarbons were carried out in Istanbul air.

In the literature the hydrocarbon pollution of cities air were based on the determination of PAHs components. In this work the amount of petroleum hydrocarbon burned or not was quantified by UVF and their components were analyzed by GC/MS and HPLC.

Additionally for the first time in literature is detected nitrophthalic acid in exhausted gas of diesel boats.

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