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# Ambient volatile organic compound (VOC) concentrations around a petrochemical complex and a petroleum refinery

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## Abstract

Air samples were collected between September 2000 and September 2001 in Izmir, Turkey at three sampling sites located around a petrochemical complex and an oil refinery to measure ambient volatile organic compound (VOC) concentrations. VOC concentrations were 4–20-fold higher than those measured at a suburban site in Izmir, Turkey. Ethylene dichloride, a leaded gasoline additive used in petroleum refining and an intermediate product of the vinyl chloride process in the petrochemical complex, was the most abundant volatile organic compound, followed by ethyl alcohol and acetone. Evaluations based on wind direction clearly indicated that ambient VOC concentrations measured were affected by the refinery and petrochemical complex emissions. VOC concentrations showed seasonal variations at all sampling sites. Concentrations were highest in summer, followed by autumn, probably due to increased evaporation of VOCs from fugitive sources as a result of higher temperatures. VOC concentrations generally increased with temperature and wind speed. Temperature and wind speed together explained 1–60% of the variability in VOC concentrations. The variability in ambient VOC concentrations that could not be explained by temperature and wind speed can be attributed to the effect of other factors (i.e. wind direction, other VOC sources).

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## 1. Introduction

Volatile organic compounds (VOCs) in the urban and industrial atmosphere mainly originate from motor vehicle exhausts and other combustion processes utilizing fossil fuels, petroleum storage and distribution, solvent usage and other industrial processes. Leaks, as well as regulated emissions, contribute to ambient concentrations of VOCs. Emission from vegetation is also an important

source of some highly reactive hydrocarbon species (Kalabokas et al., 2001). Several effects of VOCs are recognized, such as their contribution to stratospheric ozone depletion, tropospheric photochemical ozone formation, toxic and carcinogenic human health effects, and enhancement of the global greenhouse effect (Dewulf and Langenhove, 1999).

Petroleum refineries and petrochemical plants are generally large industrial installations. Their operation is associated with the emission of various organic compounds into the atmosphere, mainly originating from the production processes, the

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storage tanks and the waste areas (Kalabokas et al., 2001). In petrochemical industries, most of the organic compounds are derived from petroleum fractions, and actually from only a few basic hydrocarbons such as methane, ethane, propane, benzene, toluene, and xylene (Crosby, 1998). Petroleum refining is the physical, thermal and chemical separation of crude oil into its major distillation fractions. The primary products of the industry fall into three major categories (EPA, 1995): fuels (motor gasoline, diesel and distillate fuel oil, liquefied petroleum gas, jet fuel, residual fuel oil, kerosene and coke); finished non-fuel products (solvents, lubricating oils, greases, petroleum wax, petroleum jelly, asphalt and coke); and chemical industry feedstocks (naphtha, ethane, propane, butane, ethylene, propylene, butylenes, butadiene, benzene, toluene and xylene). Leaks, burning of fuels in process heaters and various refinery processes are the emission sources in refineries (EPA, 1995). Along with VOCs, various other pollutants (i.e. sulfur dioxide, reduced sulfur compounds, carbon monoxide, nitrogen oxides and particulate matter) are emitted from petroleum refineries (Buonicare and Davis, 1992).

Recently, atmospheric concentrations of hydrocarbons were measured at several sites around an oil refinery near the city of Corinth, Greece (Kalabokas et al., 2001). This study by Kalabokas et al. (2001) indicated that the ambient air concentrations of saturated and aromatic hydrocarbons in a large area surrounding an oil refinery were lower than the ambient air quality standards and the ambient air concentrations measured in the two large urban centers in Greece.

Izmir metropolitan city, with a population of 2 700 000 is the center of a highly industrialized area on the Aegean Sea shoreline of Turkey. There are many industries emitting high quantities of air pollutants in the area. A petroleum refinery and a connected petrochemical complex are located at approximately 60 km northwest of Izmir city center in the Aliaga region. The refinery processed  $9.14 \times 10^6$  t of crude oil in 2001 and produced  $8.42 \times 10^6$  t of petroleum products (liquefied petroleum gas, naphtha, unleaded gasoline, premium gasoline, regular gasoline, jet fuel, kerosene, gas oil, heating oil, fuel oil No 6, asphalt, lubricating

oil, extract, wax and sulfur). The petrochemical complex has a production capacity of  $1.28 \times 10^6$  t year<sup>-1</sup>. Its major products are caustic soda, hypochlorite, liquefied chlorine, polyvinyl chloride, low-density polyethylene, high-density polyethylene, ethylene glycol, polypropylene, acrylonitrile, aromatic oil, benzene, gasoline, phthalic anhydride, *p*-xylene and pure terephthalic acid. A recent study by Elbir (2002) indicated that the fuel oil use in Aliaga is high due to the petroleum refinery and petrochemical plant, accounting for 69% of high-sulfur fuel oil used in Izmir area. The petroleum refinery and petrochemical plant are the largest sources of air pollution in the region, together contributing approximately 72% to total industrial SO<sub>2</sub> emissions and 66% to overall SO<sub>2</sub> emissions (Elbir, 2002).

The objectives of this study were: (1) to measure VOC concentrations around a petrochemical complex and a petroleum refinery; (2) to investigate the effect of meteorological parameters (wind speed, wind direction and temperature) on the concentrations measured; and (3) to compare the concentrations measured with the values measured in urban and suburban sites in Izmir, Turkey and other sites around the world.

## 2. Materials and methods

### 2.1. Sampling program

Izmir is located in a basin surrounded by mountain ranges of approximately 1000–1500 m in height, with only the west end open to the Aegean Sea. The climate is Mediterranean, with warm and rainy winters, and hot and dry summers. The major air movements over the area are mainly from a northerly direction in summer. However, the predominant wind direction in winter is southerly.

Ambient air samples were collected between September 2000 and September 2001 at three selected sites around the petrochemical complex and the oil refinery, located approximately 60 km northwest of Izmir city center. Fig. 1 shows the study area and the selected sampling sites. Sampling site 1 was located at the dock of the petrochemical complex. Since the prevailing wind direction in the area is NE followed by SW, this

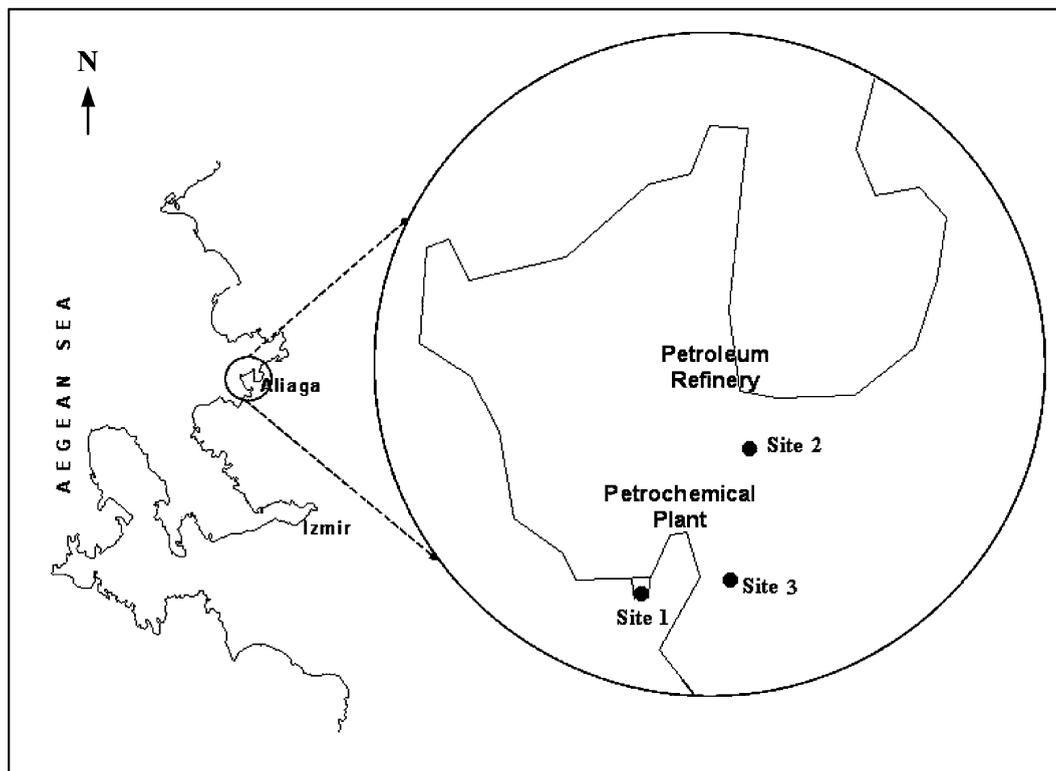


Fig. 1. General layout of the study area.

site located SW of the petrochemical plant and the oil refinery was selected to investigate the effect of both plants on ambient VOC concentrations. When the wind direction is SW, this site is expected to represent the background levels that are not affected by the petrochemical complex. Site 2 was located between the petrochemical plant and the petroleum refinery (Fig. 1). When the wind is from NE, Site 2 is affected by the refinery emissions, and when the wind is from SW this site is affected by petrochemical plant emissions. Site 3 was selected in a housing area near the petrochemical plant, located south of the petrochemical complex (Fig. 1). This site was selected to investigate the impact of the industries on housing areas when the wind blew from a northerly direction.

During the sampling program, 26 VOC samples were collected at each sampling site. All samples were collected during daytime when there was no rain. The elevation of the sample inlet was at 1.5

m above the ground level. A summary of sampling information (sampling date, sampling time, sampling flow rates) and meteorological information (wind speed, wind direction, temperature and relative humidity) is provided in Table 1. The meteorological measurements were performed using an anemometer (Testo 451) equipped with humidity and temperature sensors.

The ambient air concentrations of the following volatile organic compounds were measured: *n*-hexane; ethylene dichloride; ethyl alcohol; *n*-butyl alcohol; ethyl acetate; *n*-butyl acetate; acetone; and methyl isobutyl ketone.

## 2.2. Sampling and analysis

Ambient VOCs were collected using a sampling train consisting of a moisture trap (calcium chloride), an activated carbon tube, a rotameter, a dry gas meter and a vacuum pump. Charcoal tubes

Table 1  
Summary of sampling information

Sample number	Date	Duration (min)	Wind speed (m s <sup>-1</sup> )	Wind direction	RH (%)	T (°C)	Sampling rate (l min <sup>-1</sup> )
1	26 September 2000	161	8.5	NE		21.0	1.1
2	3 October 2000	173	4.8	NE	39	25.3	1.6
3	10 October 2000	234	6.3	NW	80	20.0	2.0
4	24 October 2000	242	3.8	NE	66	12.9	1.6
5	7 November 2000	231	1.7	SW	41	25.3	1.3
6	5 December 2000	240	2.0	N	63	15.8	1.4
7	15 December 2000	221	0.5	SW	44	15.0	1.5
8	12 January 2001	238	0.7	N	55	15.1	1.4
9	19 January 2001	285	4.7	NE	68	9.8	1.9
10	23 January 2001	229	2.8	NE	55	8.0	2.0
11	30 January 2001	229	6.2	SE	57	17.0	2.0
12	8 February 2001	228	3.7	SE	46	16.0	2.0
13	13 February 2001	250	3.0	E	71	9.0	2.3
14	28 February 2001	1658	2.7	NE	54	15.5	1.4
15	14 March 2001	1435	4.7	SW	66	18.5	2.0
16	27 March 2001	1463	5.0	SW	21	29.6	2.0
17	11 April 2001	228	4.3	S			1.1
18	19 April 2001	238	1.0	SW			1.0
19	26 April 2001	220	2.7	NW		22.0	1.1
20	4 May 2001	243	2.0	NE		24.0	1.1
21	11 May 2001	261	3.7	NE		20.0	1.0
22	6 June 2001	240	3.7	NW		30.0	0.6
23	5 July 2001	188	3.3	NE	41	32.7	1.4
24	12 July 2001	187	5.7	NE	43	31.5	0.8
25	30 July 2001	198	5.7	NE	46	32.3	1.3
26	4 September 2001	194	2.0	NE	39	30.3	0.8

Sampling information and meteorological measurements are the averages of three sampling sites.

(Dräger-NIOSH) containing 150 mg of activated carbon in two successive sections were used for sampling. The sampling flow rate ranged between 0.6 and 2.3 l min<sup>-1</sup>, and the sampling duration ranged between 130 and 1680 min. The caps of the sampling tubes were removed immediately before sampling. The sampler was attached to the sampling pump using flexible tubing (ASTM, 1988a).

After sampling, the adsorption tubes were labeled and closed with special caps to avoid contamination and desorption. The samplers were transferred to the laboratory in cold boxes. The samples were placed into tightly closed special plastic bags and stored in a freezer until they were processed.

Before analysis, contents of both sections of the adsorber tubes were placed into two different vials

and weighed, then 1.0 ml of carbon disulfide was added as the extraction solvent and they were reweighed (ASTM, 1988b). Samples were extracted in an ultrasonic bath for 15 min. They were then centrifuged for another 15 min to obtain a clear supernatant phase. The extracted samples were stored in a freezer until they were analyzed.

A micro-syringe was washed twice with the sample, washings were discarded and finally 3.0 µl of sample extract was injected into a GC. The syringe was rinsed with acetone and dried using pure air after every injection. Samples were analyzed for VOCs using a gas chromatograph (Chrompack, CP 9000) equipped with a flame ionization detector (FID). A capillary GC column (Chrompack WCOT fused silica, 50 m×0.32 mm i.d.) was used with an initial oven temperature of 40 °C, which was then raised to 120 °C at a rate

of 5 °C min<sup>-1</sup>. Chromatographic-grade pure hydrogen and air were used for the FID flame, while pure nitrogen was used as the carrier gas with a split ratio of 1:5. The GC was connected to a computer to store and evaluate the output data.

Calibration was performed using different levels of standard solutions in carbon disulfide. Liquid-phase standards were prepared by placing known volumes of chromatographic-grade pure *n*-hexane, ethylene dichloride, ethyl alcohol, *n*-butyl alcohol, ethyl acetate, *n*-butyl acetate, acetone and methyl isobutyl ketone into vials and diluting with carbon disulfide. Aliquots of 3 µl of these standard solutions were injected into the GC and run at selected conditions. Four levels of calibration standards (0.006, 0.03, 0.09 and 0.15 µl ml<sup>-1</sup>) were used to prepare the calibration curves. The concentrations of the analytes in the calibration solutions were calculated using their densities. The calibration curve was confirmed daily by analyzing a midrange standard solution. In all cases, the linear fit was good, with  $r^2 > 0.99$ .

VOCs in samples were identified by comparing their retention times to those obtained for the calibration standards under specified chromatographic conditions. The compounds identified were quantified using their peak areas in the external calibration method. The amounts of VOCs in samples were calculated using the linear regression equations obtained from the calibration curves.

### 2.3. Quality control

A midrange calibration standard containing 0.03 µl ml<sup>-1</sup> VOCs was analyzed every day to confirm the GC performance. If the percentage difference in response factor for any compound was greater than 20%, the initial calibration was assumed to be invalid and the GC was recalibrated.

Identification of individual VOCs was based on their retention times obtained from calibration runs. If the percentage difference in the retention time of any compound was greater than 5%, the compound could not be identified.

Quantifiable VOC amounts were determined from sequential injections of diluted standard solutions using a signal/noise ratio of 10. These

amounts ranged from 10 (hexane) to 26 pg (ethyl alcohol). The limit of detection based on the quantifiable VOC amounts and average sample volume (0.594 m<sup>3</sup>) ranged between 0.02 (hexane) and 0.04 µg m<sup>-3</sup> (ethyl alcohol).

The extraction solvent (carbon disulfide) was analyzed to determine if there was any contamination present. None of the compounds analyzed in this study were detected in CS<sub>2</sub>.

Blank activated carbon tubes were also extracted and analyzed as process blanks to determine if there was any contamination in the activated carbon tubes. No contamination was found in the process blanks.

Back-up sections of adsorbent tubes used in sampling were also extracted and analyzed. VOC amounts in the back-up sections were below the detection limit.

Some of the samples were analyzed in duplicate. The difference between duplicate samples was less than 5%.

Desorption efficiencies for CS<sub>2</sub> extraction are generally high and range between 86% (acetone) and 95% (ethylene dichloride and butyl acetate) (ASTM, 1988a). Therefore, VOC amounts found in samples were not corrected for desorption efficiency.

## 3. Results and discussion

### 3.1. VOC concentrations

Ethylene dichloride was the most abundant volatile organic compound, followed by ethyl alcohol and acetone (Table 2). Ethylene dichloride is a leaded gasoline additive used in petroleum refineries as a lead scavenger (Barletta et al., 2002). It is also produced in the vinyl chloride process in the petrochemical complex as an intermediate compound using ethylene and Cl<sub>2</sub>. Therefore, the high ethylene dichloride concentrations observed in this study were probably due to fugitive emissions from the petrochemical complex and the refinery.

A summary of VOC concentrations measured in this study and in a suburban area in Izmir, Turkey is presented in Table 2. VOC concentrations measured in this study around the petrochemical com-

Table 2

Summary of VOC concentrations measured in this study and at a suburban site

	VOC concentration ( $\mu\text{g m}^{-3}$ )	
	Aliaga, Izmir <sup>a</sup>	Buca, Izmir <sup>b</sup>
<i>n</i> -Hexane	4.1 ± 5.0 ( <i>n</i> =55)	1.1 ± 0.6
Ethyl alcohol	29.5 ± 34.8 ( <i>n</i> =39)	1.5 ± 2.8
Acetone	23.4 ± 30.2 ( <i>n</i> =76)	9.1 ± 6.1
Ethyl acetate	17.3 ± 34.7 ( <i>n</i> =33)	7.0 ± 4.1
<i>n</i> -Butyl alcohol	13.8 ± 23.3 ( <i>n</i> =20)	0.9 ± 0.7
Methyl isobutyl ketone	2.8 ± 3.7 ( <i>n</i> =42)	0.3 ± 0.2
Butyl acetate	6.5 ± 7.4 ( <i>n</i> =37)	1.1 ± 0.8
Ethylene dichloride	38.5 ± 59.5 ( <i>n</i> =53)	5.9 ± 4.1

Values are mean ± S.D., *n* is the number of the samples above the detection limit.

<sup>a</sup> This study, around a petrochemical complex and oil refinery.

<sup>b</sup> Cetin (2002); suburban site.

plex and oil refinery were 4–20-fold higher than those measured at the Buca suburban site. The average hexane concentration in this study ( $4.1 \mu\text{g m}^{-3}$ ) was similar to the value ( $5.1 \mu\text{g m}^{-3}$ ) measured around an oil refinery in Greece (Kalabokas et al., 2001). However, significantly higher hexane concentrations were previously measured at urban sites located near roads with dense traffic in Izmir, Turkey ( $51.1 \mu\text{g m}^{-3}$ ) (Muezzinoglu et al., 2001) and near a petrochemical complex in Korea ( $12.3 \mu\text{g m}^{-3}$ ) (Na et al., 2001). The average ethylene dichloride concentration measured in this study was higher than values previously reported. By comparison, ethylene dichloride concentrations measured by others were  $8.5 \mu\text{g m}^{-3}$  in Korea (Na et al., 2001) and  $0.25 \mu\text{g m}^{-3}$  in Karachi, Pakistan (Barletta et al., 2002).

### 3.2. Spatial and temporal variation

Total VOCs refer to sum of the concentrations of eight compounds analyzed in this study. Total VOC concentrations (average ± S.D.) were  $112.8 \pm 128.3$ ,  $73.3 \pm 102.2$  and  $64.32 \pm 70.3 \mu\text{g m}^{-3}$  for Sites 1, 2 and 3, respectively (Table 3).

The prevailing wind direction during the sampling program was NE, followed by SW (Fig. 2). The highest total VOC concentration was measured at Site 1, followed by Site 2 (Table 3). Site 1

Table 3

Summary of total VOC concentrations at the sampling sites

Site	VOC concentration ( $\mu\text{g m}^{-3}$ )				<i>n</i>
	Average	S.D.	Min.	Max.	
Site 1	112.8	128.3	15.6	485.9	25
Site 2	73.3	102.2	0.6	480.3	26
Site 3	64.3	70.3	5.1	289.6	26

had the highest average concentration because during most of the sampling program it was influenced by the combined effects of the petrochemical complex and refinery emissions due to northerly winds (Figs. 2 and 3). However, Site 2, having the second-highest average total VOC concentration, was affected by either petrochemical plant emissions (when the wind was from SW) or refinery emissions (when the wind was from NE or NW). The highest concentrations at Sites 1 and 2 were observed for northerly wind directions. The concentration at Site 3 was relatively lower than at the other sites. Site 3 was located SE of the sources investigated and the other sampling sites. Therefore, this site was relatively less affected by petrochemical plant and refinery emissions, since the wind was mostly from NE or SW during the sampling program. The highest concentrations at Site 3 were measured when the wind direction was NW (Fig. 3). The total VOC concentrations measured at Sites 1 and 3 when the wind was from SE

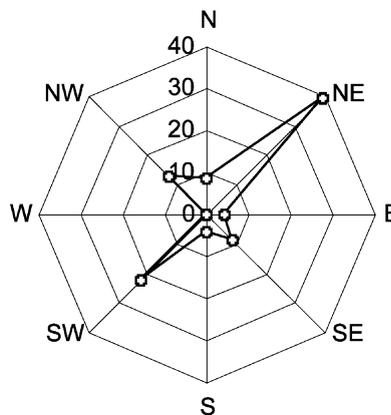


Fig. 2. Frequency (%) diagram of wind direction for the sampling program.

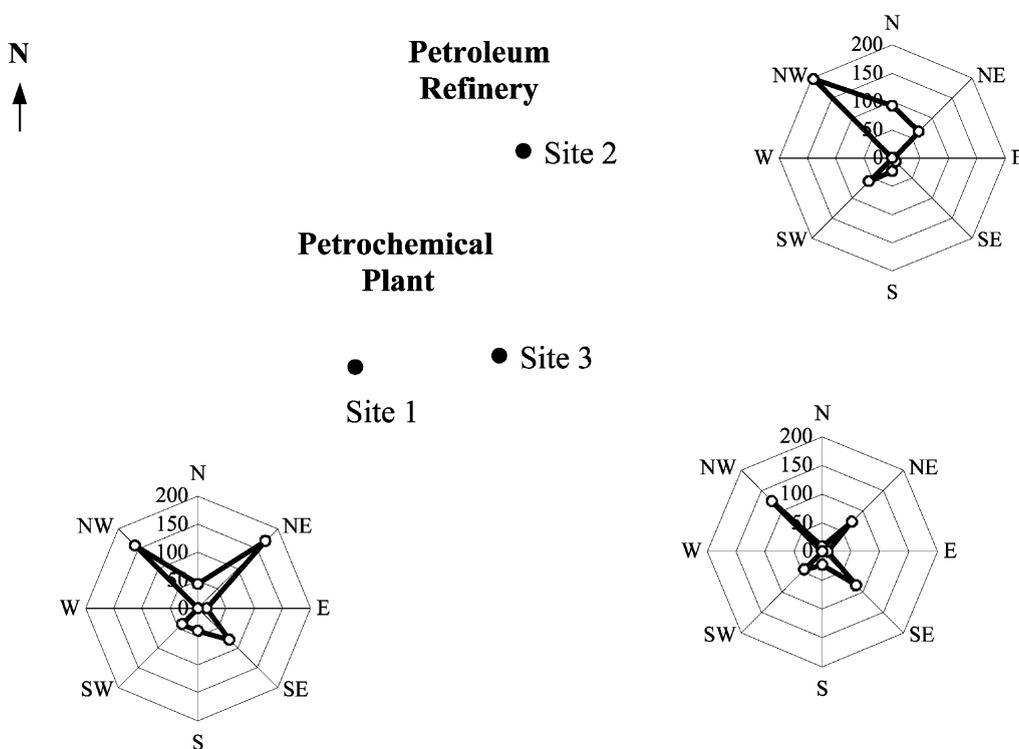


Fig. 3. Rose diagrams of total VOC concentration ( $\mu\text{g m}^{-3}$ ) for the sampling sites.

were probably affected by motor vehicle emissions from the highway located to the south of these sites.

Temporal variation of total VOCs is presented in Fig. 4. VOC concentrations showed significant seasonal variations. Total VOC concentrations were highest in summer, followed by autumn (Fig. 5). The higher concentrations measured in summer and autumn may be due in part to increased evaporation from fugitive sources as a result of higher temperatures.

### 3.3. Effect of temperature and wind speed

The total VOC concentrations for the three sampling sites generally increased with temperature (Fig. 4). The relationship between wind speed and VOC concentrations is presented in Fig. 6. Wind speed  $\geq 5 \text{ m s}^{-1}$  was classified as strong wind, while wind speed  $< 5 \text{ m s}^{-1}$  was classified as weak wind. High wind speeds generally result

in increased dispersion conditions. Therefore, a decrease in VOC concentrations may be expected during periods with high wind speed. However, when wind speed was  $\geq 5 \text{ m s}^{-1}$ , total VOC concentrations were relatively higher for all sites (Fig. 6). It should be noted that most of the strong wind conditions corresponded to winds from a northerly direction, carrying emissions from the two major sources to the sampling sites (Table 1, Fig. 2). Thus, the relatively higher concentrations observed in this study for wind  $\geq 5 \text{ m}^{-1}$  were probably due to the wind direction.

The relationship between individual VOC concentrations and meteorological parameters (temperature and wind speed) was further investigated using multiple linear regression analysis:

$$C = a_0 + a_1T + a_2WS \quad (1)$$

where  $C$  is the VOC concentration ( $\mu\text{g m}^{-3}$ ),  $T$  is the temperature ( $^{\circ}\text{C}$ ),  $WS$  is the wind speed (m

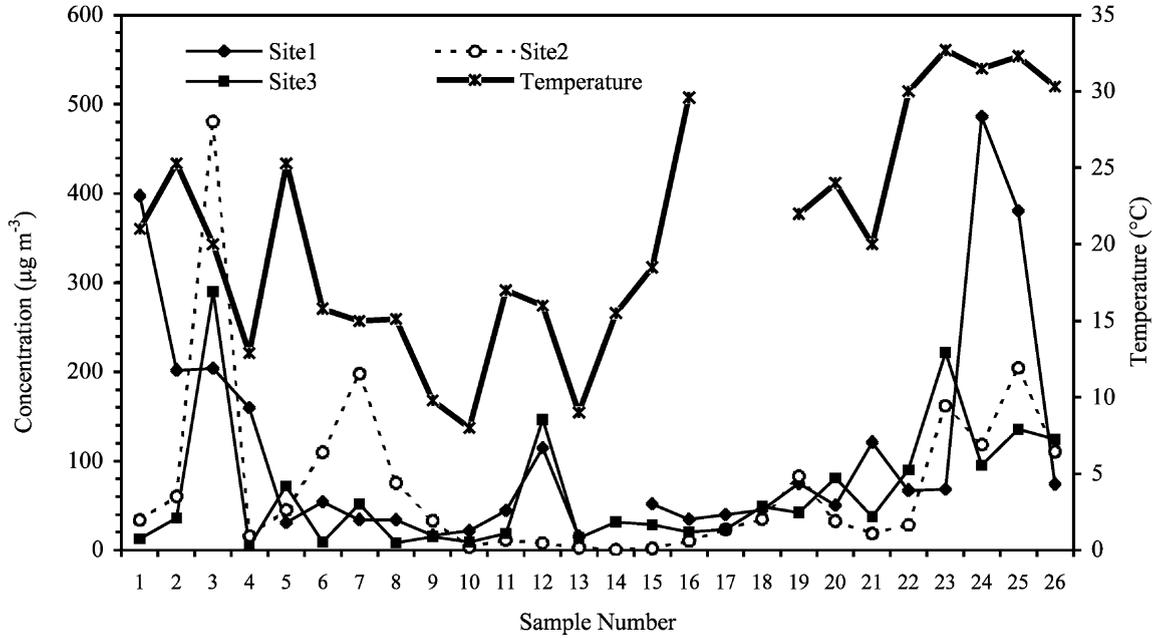


Fig. 4. Variation of total VOCs and temperature during the sampling period.

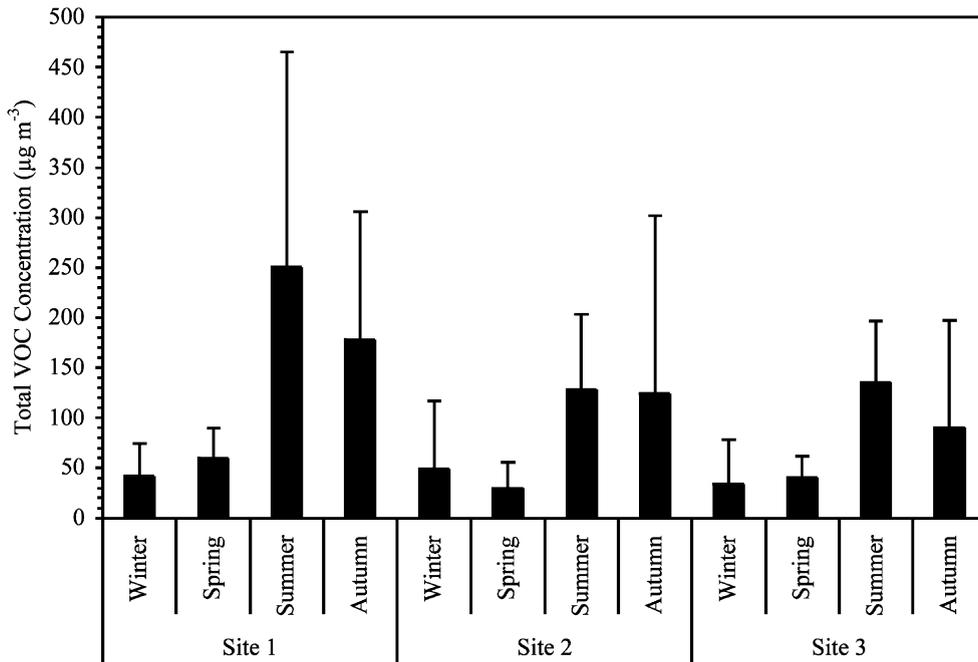


Fig. 5. Seasonal variation of VOC concentrations at the sampling sites.

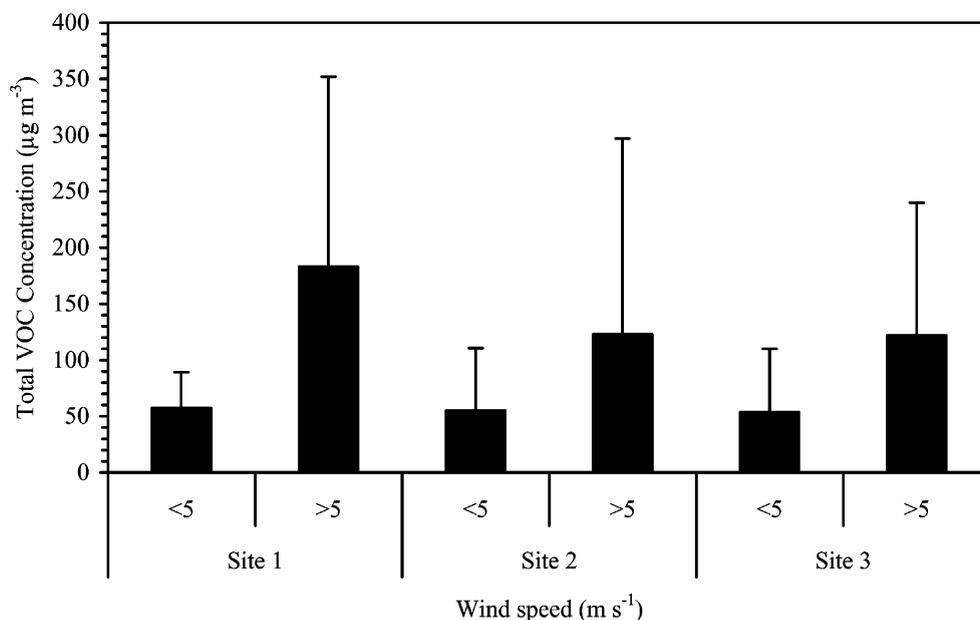


Fig. 6. Total VOC concentrations for the sampling sites at different wind speed ranges.

s<sup>-1</sup>), and  $a_0$ ,  $a_1$  and  $a_2$  are fitting parameters. Generally,  $a_1$  and  $a_2$  were positive, indicating that VOC concentrations increased with temperature and wind speed. Temperature and wind speed together explained 1–60% of the variability in VOC concentrations. Temperature accounted for 1–48% of the variability, while wind speed accounted for 1–29% of the variability in VOC concentrations. These results indicated that emissions might have increased as a result of VOC evaporation from fugitive sources during periods with relatively higher temperatures. The variability in ambient VOC concentrations that could not be explained by temperature and wind speed can be attributed to the effect of other factors (i.e. wind direction, other VOC sources).

In summary, VOC concentrations measured around a petroleum industrial site were generally higher than values measured at urban and suburban sites in Izmir, Turkey and at other sites around the world. Evaluations based on wind direction indicated that the refinery and petrochemical complex emissions affected the ambient VOC concentrations measured. VOC concentrations generally

increased with temperature and wind speed. The highest concentrations were measured in summer, followed by autumn, probably due to the increased evaporation of VOCs as a result of higher temperatures.

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