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Sulfur Dioxide (SO_2) Accumulation in Soil and Plant's Leaves around an Oil Refinery: A Case Study from Saudi Arabia

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Abstract: The accumulative levels of SO_2 in soil and plant's leaves around an oil refinery were monitored. Four different sites around the refinery area were chosen; west, south east, north east and the northern side. The refinery southern side was not accessible. In addition to the soil samples, leaves samples of the dominant plants species *Myoporum pictum* were randomly collected from all sites. Highly significant levels of sulfate were found in soil and plant leaves samples at all sites compared to the control. The highest level of sulfate in soil (9,000 ± 1200 µg g^{-1}) and plant's leaves (65,774 ± 320 µg g^{-1}) were found in the southern east side. This high content of sulfate indicates high levels of air contamination with SO_2 around the refinery which negatively effects the environment and public health at this populated area.

Key words: Air pollution, sulfur dioxide, soil and plants, oil refinery, Saudi Arabia

INTRODUCTION

Environmental pollution resulting from oil refineries is a worldwide concern. Petroleum hydrocarbon wastes leaked, emitted or even buried can cause severe damage to the biological system. A polluted area is characterized by; the high levels of emitted pollutants and organic compounds into the air, the high contamination levels of soil, plants and ground water and/or the possible movement of these contaminated matters to other areas[1]. Part of the complexity of this problem is the conflict between the economical benefits of these refineries and its hazards to the public health and the surrounding environment. Air pollution usually causes severe negative effects on biota and serious health risks on the public living in the surrounding area[2,3].

Jeddah Oil Refinery is one of seven petrochemical refineries in Saudi Arabia. It was established in the late 1960s in a non residential area[4]. Later on, this area has become crowded and highly populated.

Over all, sulfur oxides (SO_x) represent the largest amount of discharged gases into the air beside Nitrogen oxides (NO_x) around refineries[5]. Heavy metals (e. g., Cadmium, Lead, Copper and Zinc) and other pollutants (e. g., polycyclic aromatic hydrocarbons and Toluene and Phenol) are also emitted[5].

Sulfur dioxide (SO_2), with its specific smell, is the most dominant gas associated with air pollution emitted by oil refineries[6]. It accumulates at high levels in the soil and higher plants in the surrounding areas. Sulfur dioxide (SO_2) is usually trapped in the lower atmospheric air layer. This close vicinity allows it to reach the ground and to accumulate on the surface soil easily. Rainfall, humidity and hot climate increase the SO_2 solubility in air and its precipitation accelerates its accumulation in soil and plants based on air inversion[7-9]. Therefore, monitoring its levels in soil and plants around refineries has been used as indicators of air pollution in many studies[1,8,10-12].

The heath hazards of SO_2 gas to the public living around these refineries result from long term inhalation exposure to the gas which is believed to be responsible for many diseases such as allergic rhinitis, lung inflammations, lung cancer[1,13,14]. Many studies confirmed the association of respiratory and cardiovascular diseases with long term exposure to air polluted with SO_2[3,13-15]. Our major concern is directed towards the SO_2 cycle and its involvement in the food chain. This can be easily achieved through monitoring the accumulated level of SO_2 in the soil and plant's organs around the refinery areas. Other studies revealed dramatic changes in soil pH moving towards acidity and causing severe damages to microbial community which affects the soil fertility[16,17].

Plants in polluted areas are more affected by SO_2 than other living organisms including humans[18]. Plant's leaves take up SO_2 molecules through two

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pathways, their open stomata and through their roots[19]. A previous study has shown that sulfate compounds accumulate more in old plant leaves than the younger ones due to long term exposure, at the same time some plants species appear to be more sensitive than others, although all plants species are sensitive to SO_2 gas[19]. Another study conducted in Oman, used plant's leaves as biomarkers for SO_2 level around Muscat Oil Refinery[8].

The present study is designed to assess the accumulative levels of sulfate in soil and plant's leaves at four different sites around an oil refinery and to evaluate the environmental conditions for the area under study.

MATERIALS AND METHODS

The study site: The study was carried out from March to May 2006, on the residential area, approximately 100 m away from the Jeddah Oil Refinery. Four sides were chosen; west, south east, north east and the northern side. The southern area of the refinery was not accessible. These sites were carefully selected to reflect the actual environmental state and to cover all inhabited areas considering the wind direction factor. Environmental conditions and flora were almost similar at all study sites. No interference with other source of pollution was considered. Soil and plant's leaves of *Myoporum pictum* tree samples were collected from all sites. Control samples were obtained from pollution-free area ~ 50 km outside the city limit.

Samples collection: Our primary survey showed different plants species were present. *Myoporum pictum* tree, commonly known as Bazrroii, was the most dominant species and was found at all study sites. Therefore, this species was studied to eliminate the species differences factor. Fresh leaves of *Myoporum pictum* tree of same height were collected in marked bags. Due to the low annual rate of precipitation in this area, soil samples were mainly collected from the ground surface up to 10 cm depth only. All samples were placed into marked containers.

Samples Preparation for Sulfate Analysis

Plant's leaves: After gentle brushing, the leaves were washed with cold tap water and then by distilled water. Leaves samples were dried at 80 ^{o} C for 24 h and then gently grinded. From each sample, 1 g of fine grind dried leaves was digested with concentrated HNO_3 and prechloric acid in a fumigation chamber. The total sulfate concentration was determined according to Chapman and Pratt[20].

Soil: The sulfate was extracted using a 1:1 mixture of acetic acid 0.24M and ammonium acetate 0.05M. The soil sample (10g) was treated with 25 ml of the extracting solution and then shaken for 30 min in a centrifuge (200 oscillations per min). Activated charcoal was then added (0.25g) and the mixture shaken for 3 min. The samples were then filtered using sulfate free filter paper. Filtrates were then diluted and Sulfa Ver® 4 pillow powder (HACH Company, USA) was added to the diluted samples. Concentrations of the sulfate were measured at 450 nm using APEL spectrophotometer (PD- 303 UV model with APEL software 2.0) according to Chaudry and Cornfield[21].

Statistical analysis: Data statistical analysis was preformed using SPSS 11.0 for Windows. All descriptive data were represented as means ± SE. The *t*-test was used to evaluate the concentration levels of sulfate in soil and plant's leaves compared to the controls. Analysis of variance (ANOVA) with Bonferroni correction test were preformed for means multiple comparisons among sites. Standard liner curve and the equation were obtained using a liner regression model.

RESULTS AND DISCUSSION

The map of the studied area is illustrated in Fig. 1. The locations of sampling sites around Jeddah Oil Refinery covered all accessible sides that are subjected to the discharged gases. Table 1 shows the levels of SO_2 accumulated in soil and *Myoporum pictum* leaves collected from all studied areas versus control samples.

Table 1: Overall levels of Sulfate content in soil and plant's leaves collected from all study sites

Location	Sulfate content in leaves ($\mu g\ g^{-1}$)	Sulfate content in soil ($\mu g\ g^{-1}$)
West side	41,109 ± 240 [1]	5,312 ± 562 [1,2]
Southern east side	65,774 ± 390 [1,2]	9,000 ± 1200 [1,2]
Northern east side	50,701 ± 270 [1,2]	4,115 ± 713 [1,2]
North side	43,850 ± 185 [1]	3,200 ± 159 [1]
Control	10,000 ± 102	1,120 ± 220

[1]t-test significant differences with control ($P < 0.05$)
[2]ANOVA with multiple comparison tests for significant differences among sites ($P < 0.05$)

As shown in Table 1, the sulfate concentration levels in both soil and plant's leaves samples from all study sites were higher than the control ($P < 0.05$).

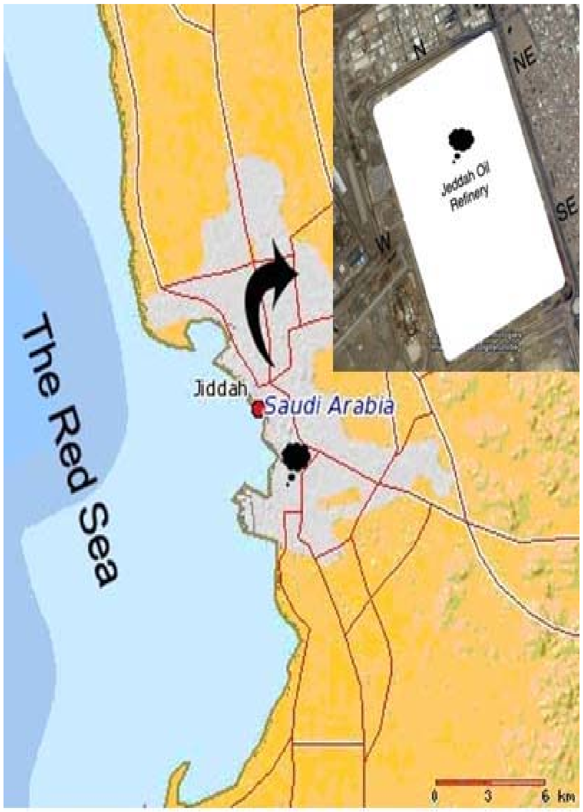

Fig. 1: Map of the study area shows the four sampling sides around Jeddah Oil Refinery

Analysis of variance with multiple comparison tests also showed significant differences in soil and plant's leaves among most sites ($P < 0.05$). It is clear from the Table that the soil and plant's leaves samples collected from the south eastern side of the refinery showed the highest sulfate levels. The sulfate concentration in the plant's leaves samples ranged between 65,774 ± 390 µg g^{-1} (South east) and 41,109 ± 240 µg g^{-1} (West). While soil samples varied between 9,000 ± 1200 µg g^{-1} (South east) and 3,200 ± 159 µg g^{-1} (North). Morphologically, *Myoporum pictum* leaves possessed miner visible injuries that can be summarized as; leaf shortness and yellowness. No major injuries were noticed.

Most published literature concerning gaseous air pollution focused on Sulfur dioxide SO_2 contamination. The present study is an applied local field evaluation of the air pollution around an oil refinery. The accumulative levels of SO_2 in soil and plants have been used as field indicators reflecting the degree of air pollution in the studied areas. Jeddah Oil Refinery, the oldest among Saudi Refineries, was built outside the city limits. Over the years, Jeddah city population has increased rapidly and the refinery is now in the middle of a crowded area, creating the most serious source of air pollution in the city. Consequently, the potential health hazards to the public resulting from the pollutant gases emitted daily have dramatically increased over time. Sulfur dioxide (SO_2) is the most abundant, most widely studied air pollutant discharged from oil refineries[6]. In the case of Jeddah Oil Refinery, this gas is emitted continuously 24 hours on a daily bases affecting the public health and the environment in the nearby area. Sulfur dioxide (SO_2) gas is rapidly dissolved in the atmospheric water vapor forming acid rain that accumulates in air, soil and plants around refineries[1,16]. A major warning from the threat to public heath and to the environment resulting from Jeddah Oil Refinery has been early issued[22]. The threat is focused on two major concerns, the gaseous emissions into the air and the crude oil and hydrocarbons wastes affecting marine waters. Over the past years, many investigators have demonstrated the negative impacts of Sulfur dioxide (SO_2) on human health[13,23] and on biota[8,11,24]. However, It is not easy to study the effects and the levels of gaseous air pollutants like SO_2 in the different environmental components such as air, soil and plants. The complexity results from the fact that this gas actively reacts with some ions such as Mg, K, Ca found in the air or soil. Such reactions usually form different sulfate compounds[16,25].

Higher plants have high sensitivity to atmospheric SO_2 than other living organisms[18]. Basically, sulfate compounds can easily reach the plant leaves through either the stomata or the roots. With long term exposure, they accumulate usually in the old leaves more than the younger ones[19]. Plants respond to SO_2 exposure either by accumulating more sulfate in their internal tissues or by showing visible injuries[12]. Many physiological changes associated with SO_2 stress have been reported in plants including, reduction of stem length and height, decrease in leaf size and decline of the phytosynthetically active tissues[26]. The miner visible injuries observed on *Myoporum pictum* leaves in the present study may suggest that this species has high tolerance to extreme sulfate levels[10]. Hence, pervious studies have successfully used the accumulated levels of SO_2 in plants leaves as a useful tool for air pollution state around oil refineries[8,12,26].

The present study has shown that high levels of SO_2 gas derivatives accumulating in soil and plants around an oil refinery. These results are supported by previous findings conducted on plant's leaves around Muscat Oil Refinery, Oman[8]. The recorded levels of sulfate in *Myoporum pictum* leaves around Jeddah Oil Refinery exceeded the levels reported by Abdul-Wahab and Yaghi[8] in other plant species around Muscat Oil Refinery. Many reasons could explain the variation between the two refineries: Jeddah Refinery is a very old plant, discharging gases for almost half a century. The highly hot and humid climate of Jeddah city accelerates the rate of SO_2 accumulation in air, soil and plants. Different plant species studied showed different sulfate levels. The present study showed that the soil and leaves samples at the southern east side of the refinery expressed the highest content of sulfate. This could be explained by the fact that wind direction drives smoke towards the east most of the year.

Other modern Saudi refineries do not experience the same problem of SO_2 pollution as the refinery under study. In these refineries, the produced SO_2 is treated. For instant, Yanbu Refinery is supported by a seawater flue gas desulphurization system (SWFGD) provided by ALSTOM Co. This system cuts SO_2 emission by up to 90% using seawater as the absorbent to neutralize SO_2 transform it into sulfate, which is a natural compound of seawater[27].

Some investigators have suggested few protective steps such as health awareness and intensive safety training programs to the Saudi refineries employees[4]. These precautionary steps can not solve the SO_2 emission problem, nor reduce the damage to public health and to the environment.

CONCLUSION

The present study emphasized the major threat of sulfur dioxide (SO_2) discharged by Jeddah Oil Refinery to public health and to the environment. Therefore, we recommend that the emission of sulfur dioxide (SO_2) should be reduced and safety training programs and environmental awareness to be intensified.

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