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ATMOSPHERIC CONCENTRATIONS OF INORGANIC POLLUTANTS (NO_2, SO_2 AND OZONE) IN ESKİŞEHİR: SPATIAL AND VERTICAL VARIATIONS, WEEKDAY-WEEKEND DIFFERENCES

Özlem ÖZDEN ÜZMEZ *

Department of Environmental Engineering, Faculty of Engineering, Anadolu University, Eskişehir, Turkey

ABSTRACT

Atmospheric concentrations of inorganic pollutants (NO_2, SO_2 and ozone) were measured at three different sampling sites in Eskişehir. Samples were collected between 25th of March and 8th of April 2016 by using passive sampling method. During two-week sampling study, each week was divided to three sub-periods to investigate weekday and weekend differences of the pollutant concentrations. One-time vehicle counts during morning, noon and evening hours for both weekday and weekend periods were inspected. The highest NO_2 concentrations were measured at the sampling site having urban, traffic and residential characteristics. Positive relationship between NO_2 concentrations and traffic density was observed. Reverse trend was determined for ozone. Ozone concentrations were higher at the sampling site far from the city center. Spatial variations of SO_2 concentrations were similar to those of NO_2. Weekday NO_2 concentrations were higher than weekend concentrations while higher ozone concentrations were observed in weekend. Weekend and weekday SO_2 concentrations were quite similar at all sampling sites. Beside spatial variations, vertical variations of each pollutant were also investigated. NO_2 concentrations decreased with the elevation and reverse case was observed for ozone with increasing concentrations at the same elevation range. SO_2 concentrations increased with the elevation due to chimney gas emissions.

Keywords: NO_2, SO_2, O_3, Passive sampling, Urban air quality

1. INTRODUCTION

Air pollution is one of the most important environmental problems. Especially, increasing of population density, industrial activities and use of motor vehicles in urban environments leads to increase in the levels of atmospheric pollutants. Atmospheric concentrations of the pollutants are important in terms of human and environment health. Inorganic gas pollutants such as nitrogen dioxide (NO_2), sulphur dioxide (SO_2) and ozone (O_3) are among the most important atmospheric pollutants. Among these, SO_2 and NO_2 are conventional air pollutants. The main sources of NO_2 and SO_2 in the atmosphere are road traffic, domestic heating, and industrial emissions [1-3]. Ozone is a secondary pollutant [4-6]. Emissions of nitrogen oxides (NO_x) and Volatile Organic Compounds (VOCs) from both man-made and natural sources react in the presence of sunlight to form ground-level ozone in the atmosphere [7-9]. It can be harmful to human health and plant life and can damage materials [10, 11].

Passive sampling offers several advantages compared to other sampling methods. Unlike active (pumped) sampling, passive samplers require no electricity, are portable, inexpensive and simple to use (no pump operation or calibration). Also, they are suitable for personal monitoring, indoor and outdoor air analysis, offer indication of average pollution levels over time periods of 8 hours to weeks/months [12-17]. Due to these advantages, use of passive samplers in many air pollution sampling studies have increased reasonably in recent years [18-24].

When the determination of air quality of an urban area is aimed, sampling studies are performed at many sites with different characteristics such as urban, suburban, rural, urban background. In this study,

*Corresponding Author: oozden@anadolu.edu.tr
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ambient air quality of Eskişehir that is a city located in northwestern Turkey was determined by using passive sampling method. Specific objectives of this study were: (1) to determine atmospheric NO_2, SO_2 and O_3 concentrations at three different sampling sites that have (i) urban, traffic and residential, (ii) urban, residential, (iii) suburban, residential characteristics, (2) to investigate weekday-weekend differences of the pollutant concentrations, (3) to determine the vertical variations of the pollutant concentrations at the sampling sites.

2. EXPERIMENTAL

2.1. Study Area and Sampling Program

In this study, ambient NO_2, SO_2 and O_3 concentrations were measured in the urban atmosphere of Eskişehir by using passive sampling method. Passive samples were collected from three sampling sites located in the different regions of the city (Figure 1).

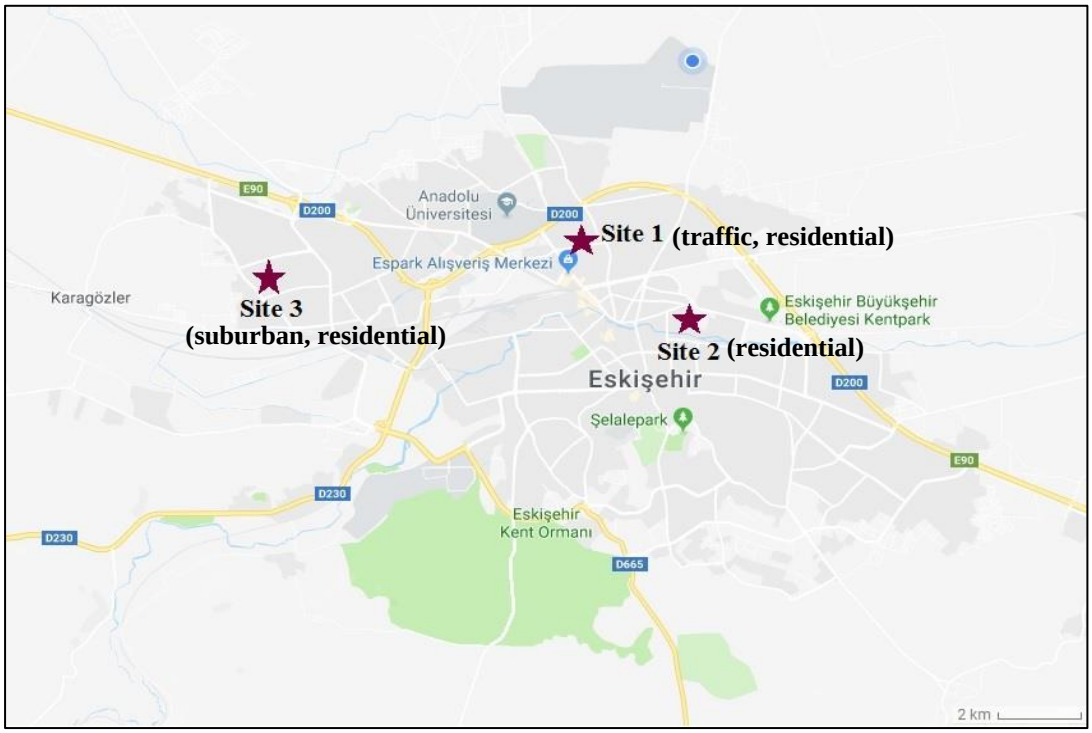

Figure 1. Locations of the sampling sites

Sampling sites were selected due to their locations and characteristics. *Sampling Site 1* can be characterized as urban, traffic and residential. It is located in the city center and has a high traffic density. *Sampling Site 2* has only urban and residential characteristic (it is not in the city center and does not have dense traffic). *Sampling Site 3* is located approximately 10 km far from the city center. It can be classified as suburban and residential since there are mostly detached houses in this region.

Sampling studies were carried out continuously between 25th of March and 8th of April 2016 at all sampling sites. During two-week period, each week was divided to three sub-periods such as Friday-Monday (weekend), Monday-Wednesday (weekday) and Wednesday-Friday (weekday). So, weekday and weekend samplings were carried out separately at all sampling sites. One-time vehicle counts for six classes during morning (08:00-09:00), noon (12:00 -13:00) and evening (18:00-19:00) hours for both weekday and weekend samplings were inspected by using a hand-made equipment for all the sampling sites. Simply, small tally counters which were assigned for different vehicle types were fixed to a styrofoam platform. A person sitting nearby to road were manually pushing the corresponding button of

the counter for each passing vehicle type. The vehicle count data was given in Figure 2. As shown in Figure 2, for both weekday and weekend and so for whole week, the highest vehicle count was obtained for Sampling Site 1, while the lowest vehicle number was counted at Sampling Site 3.

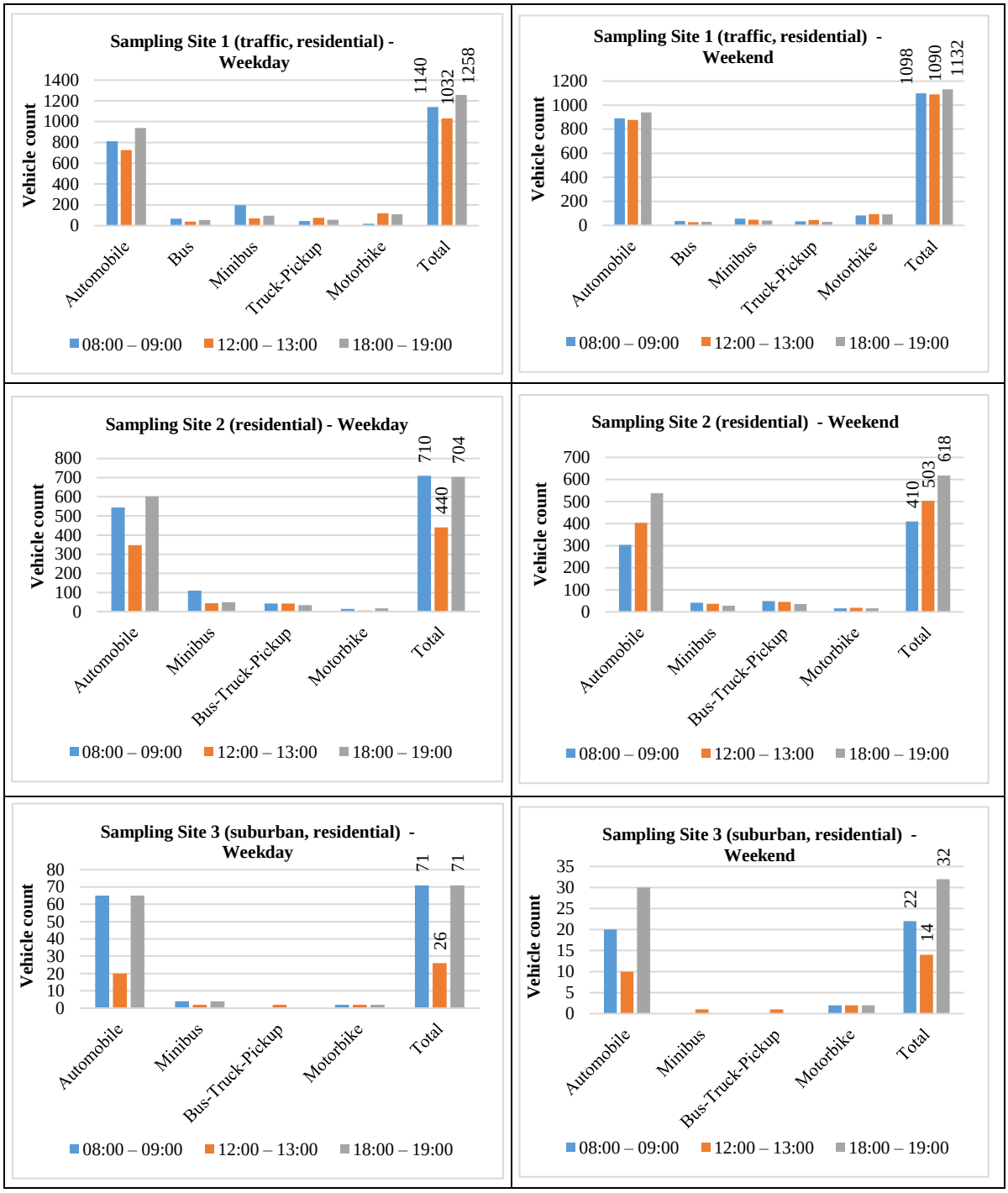

Figure 2. Weekday and weekend traffic counts obtained for the sampling sites

Within a city there are not only variable concentrations of pollutants at different sites but there is also a vertical variation. So, vertical variations of the pollutants were also investigated by placing the passive samplers at different floors of the buildings at the sampling sites. At Sampling Site 1 and 2, the samplers were placed to the apartments while the building was a detached house at Sampling Site 3. Passive

samplers were placed at 7 m (1st floor), 13 m (3th floor), 25 m (7th floor) and 31 m (9th floor) from the ground at Sampling Site 1, while the elevations were 7 m (1st floor), 13 m (3th floor) and 19 m (5th floor) at Sampling Site 2. The height of the point where the passive samplers were placed was 7 m at Sampling Site 3.

Since the first floors of the buildings are the closest to the ground level and so affected from local ground level activities such as traffic, divided samplings such as weekend and weekday samplings were carried out at only first floors while the samplings at other floors were performed during one-week period of the two-week sampling.

Three replicate passive samplers and also one blank sampler were prepared for each floor at each sampling site and 100 passive samplers were used in total.

2.2. Preparation of the Passive Samplers and Analyses of the Samples

In this study, passive samplers developed and validated by Anadolu University Environmental Engineering Department Air Quality Research Group were used for the measurements. The passive samplers have been used in several studies [21, 24-28]. NO_2 and SO_2 were collected in the same sampler, while different sampler was used for ozone. All the samplers have same dimensions with 2.5 cm length and 2.0 cm inner diameter. The main parts of the passive samplers are also same: (1) sampler body, (2) stainless steel mesh barrier, (3) close cap, (4) filter paper impregnated with specific solution and (5) fixer ring. The passive sampler body was manufactured from polytetrafluoroethylene (PTFE) for NO_2-SO_2 and delrin for ozone. For collecting medium, Whatman GF/A glass-fiber filter papers impregnated with 20% TEA (Triethanolamine) aqueous solution for NO_2-SO_2 and aqueous solution containing 1% $NaNO_2$, 2% Na_2CO_3 and 2% glycerol for ozone were used. The picture of the NO_2-SO_2 passive sampler and its parts was given as an example in Figure 3.

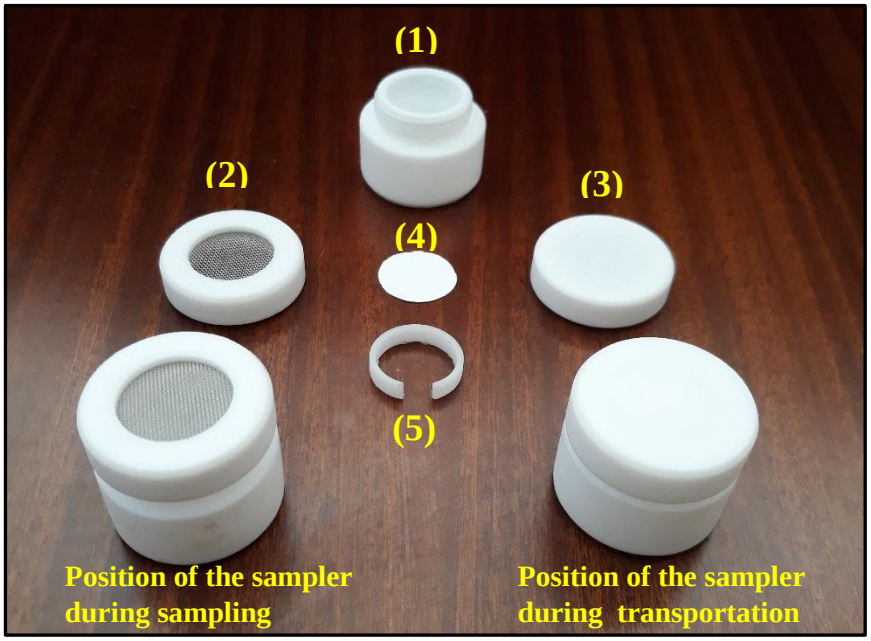

Figure 3. Parts of the tailor-made passive sampler

For each sampler, impregnated filter paper was dried and placed at the bottom of the sampler and fixed with the ring. The inlet end was closed with a plastic cap. To minimize the turbulence effect of wind inside each sampler, a stainless steel mesh barrier was placed at the open end during sampling, and the barrier was replaced with the close cap during the transportation of the samplers. Also, passive samplers were placed

in the shelters during sampling period to minimize the negative effects of some meteorological parameters such as wind velocity, rain, snow etc. and the shelters were hung in the balconies of the buildings. Before the analyses of the samples, extraction procedure was carried out. Extractions of NO_2-SO_2 filter papers were performed in 10 mL ultra-pure water (Milli Q) + 20 µL 35% H_2O_2 (Merck) solution and ozone filter papers were extracted in 10 mL ultra-pure water (Milli Q) during 15 minutes. Analyses of the all samples were carried out by Dionex ICS-1100 ion chromatograph. Concentrations of the measured pollutants were determined based on Fick's first law of molecular diffusion [29].

2.3. Quality Control/Quality Assurance (QC/QA) Parameters

During the analysis procedure, NO_2, SO_2 and ozone were analyzed as nitrite (NO_2^-), sulphate (SO_4^{2-}) and nitrate (NO_3^-), respectively. Instrumental detection limit (LOD) was calculated by performing analysis of the second lowest calibration standard for ten times and multiplying the standard deviation of the measurements by three. LOD values for NO_2^-, NO_3^- and SO_4^{2-} were 0.20, 0.18 and 0.21 µg, respectively. Precisions of the analyses were determined as coefficient of variance (CV, %) by using six replicate measurements of the intermediate standard solution. CV values were obtained as 4.5%, 7.0% and 3.7% for NO_2^-, NO_3^- and SO_4^{2-}, respectively. Field blanks were subtracted from the measurement results. Average blank values for NO_2^-, NO_3^- and SO_4^{2-} were 0.25±0.06 µg, 0.20±0.04 µg, 0.23±0.07 µg, respectively. Extensive validation results of the passive samplers were reported in other studies [24-28, 30].

3. RESULTS and DISCUSSION

3.1. Spatial Concentrations of the Pollutants

Average concentrations of the pollutants measured at three sampling sites (at the first floors of the buildings) were shown in Figure 4. Two-week average concentration of NO_2 was 66.66±9.78 µg m^{-3} at Sampling Site 1; 50.23±10.47 µg m^{-3} at Sampling Site 2 and 25.18±10.16 µg m^{-3} at Sampling Site 3. The highest NO_2 concentrations were measured at Sampling Site 1 that has urban, traffic and residential characteristics. Also, the trend in NO_2 concentrations measured in the sampling sites was similar to trend in vehicle count data obtained for each sampling site (Figure 2). This result is expected since the major ambient source of NO_2 is traffic. The relationship between NO_2 concentrations and vehicle number was also shown in Figure 5. As seen in Figure 5, the relationship between two parameters was quite high with R^2=0.81.

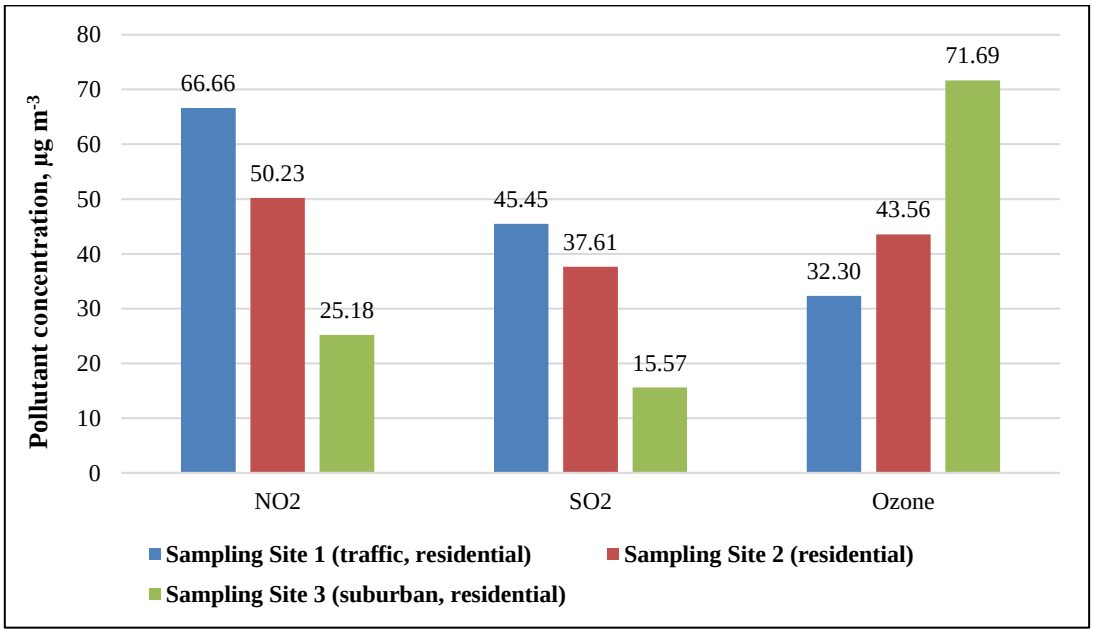

Figure 4. Average pollutant concentrations measured at the sampling sites

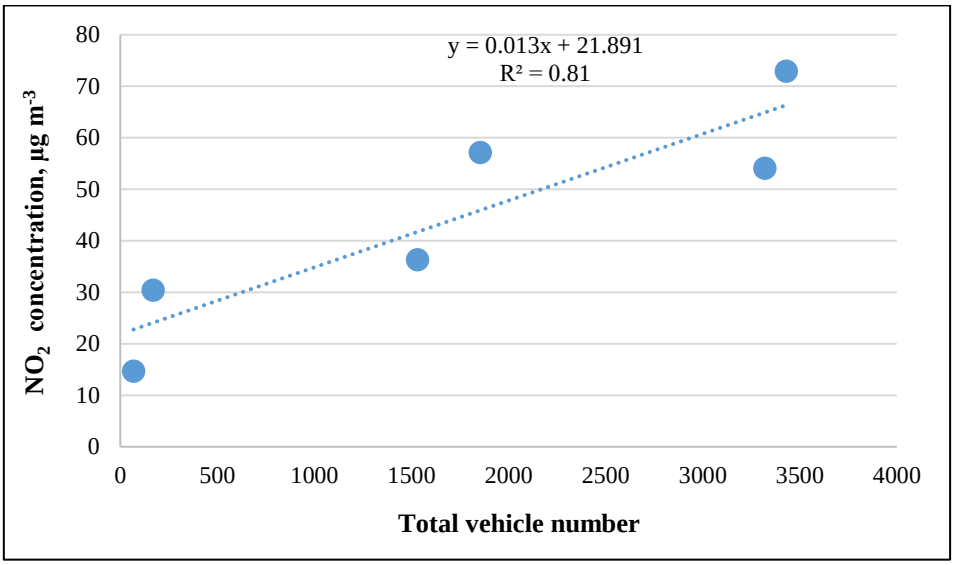

Figure 5. Relationship between NO_2 concentrations and total vehicle number

Spatial variation of SO_2 concentrations was similar to those of NO_2 pollutant. Two-week average concentration of SO_2 was 45.45±8.90 µg m^{-3} at Sampling Site 1; 37.61±5.87 µg m^{-3} at Sampling Site 2 and 15.57±3.79 µg m^{-3} at Sampling Site 3. In general, SO_2 is a seasonal pollutant and the major source of ambient SO_2 is combustion of sulphur-containing fossil fuel. During the sampling period, it was seen that domestic heating was also applied partially in the regions where sampling sites were located. In Figure 6, both variations of SO_2 concentrations and ambient air temperatures obtained during two weeks were given for Sampling Site 1. There was a reverse relationship between SO_2 concentrations and ambient temperatures. During the dates with low temperatures, higher SO_2 concentrations were measured. One of the major reasons for this reverse relationship was thought to be application of domestic heating activities in colder days. In Eskişehir, both natural gas and coal are used for domestic heating and especially at Sampling Site 1 and 2, both fuel type is used for the heating purpose. At Sampling Site 3, natural gas is used mainly. So, SO_2 concentrations were higher at Sampling Site 1 and 2 compared to Sampling Site 3. Higher SO_2 levels were also associated with population density. Since Sampling Site 1 is located in much more populated area compared to other sites, the highest average SO_2 concentration was measured at Sampling Site 1 due to the highest population.

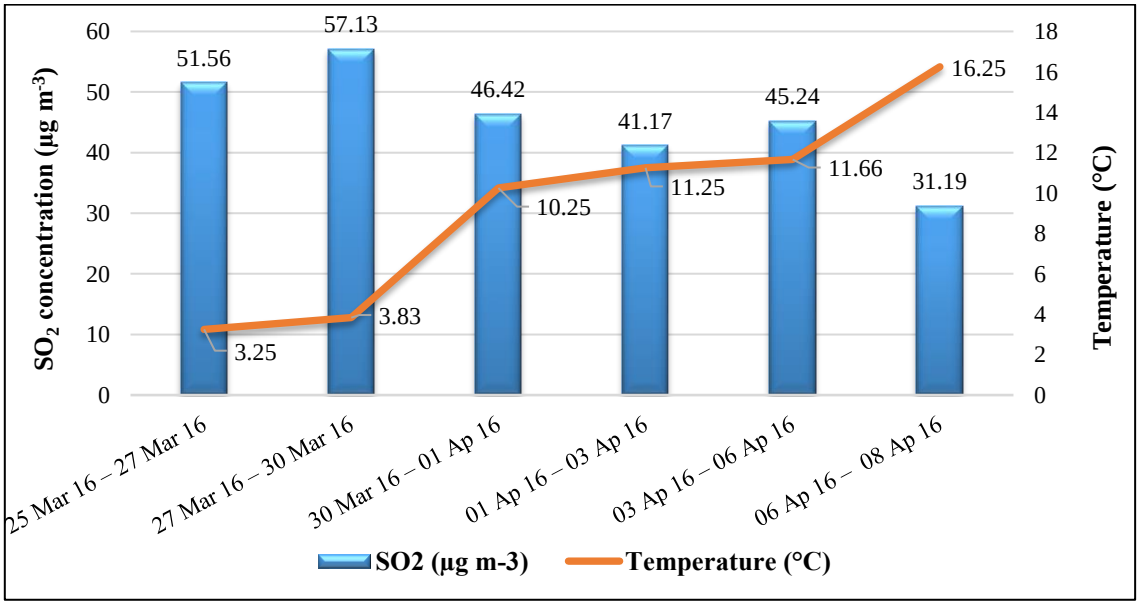

Figure 6. Simultaneous trends in SO_2 concentrations and ambient air temperatures for Sampling Site 1

Two-week average concentration of ozone was 32.30±12.40 μg m^{-3} at Sampling Site 1; 43.56±10.92 μg m^{-3} at Sampling Site 2 and 71.69±6.11 μg m^{-3} at Sampling Site 3. Contrary to NO_2, ozone concentrations were higher at the sampling site far from the city center (Sampling Site 3) (Figure 4). Low ozone concentrations in the city center are related to the high NO_2 levels. This is generally caused by the depletion of ozone by NO in the center, especially in the areas with high traffic density. Reverse relationship between NO_2 and ozone concentrations measured at the sampling sites can also be seen from Figure 4.

3.2. Weekday-Weekend Differences of the Pollutant Concentrations

Weekday and weekend concentrations of the pollutants were measured separately during the study. Table 1 shows weekday and weekend concentrations of the pollutants measured at all sampling sites.

Table 1. Weekday -weekend concentrations of NO_2, SO_2 and ozone (μg m^{-3}) at the sampling sites

Sampling Site	Pollutant	Weekday	Weekend
1	NO_2	72.93±5.88	54.11±2.53
(traffic, residential)	SO_2	45.00±10.64	46.37±7.35
	Ozone	24.90±2.44	47.12±9.73
2	NO_2	57.19±4.88	36.33±2.58
(residential)	SO_2	36.82±5.90	39.18±7.77
	Ozone	37.14±5.80	56.40±2.31
3	NO_2	30.41±9.25	14.73±3.53
(suburban, residential)	SO_2	15.40±5.34	15.92±5.00
	Ozone	68.38±2.93	78.33±3.11

In urban areas, traffic density and also different human activities vary between weekdays and weekends. So, the difference between the weekday and weekend concentrations of the pollutants allows us to get an idea about the contribution of the different sources on the pollutant levels. NO_2 (one of the ozone precursors) concentrations were lower in weekends than those found in weekdays at all sampling sites. The lower NO_2 concentrations in weekends may be due to lower traffic density in weekends since most of the people doesn't work (there is no rush hour traffic during weekend period) and also weekends are school holiday periods and so school transport services are not active in the roads. High weekday concentrations of NO_2 are also related with low weekday ozone concentrations since NO_2 is one of the important ozone precursors [26]. As seen from Table 1, ozone concentrations were higher in weekends. This phenomenon is known as "weekend effect" [11, 31-33]. "Weekend effect" occurs when ozone concentrations are observed higher in weekend than that of weekday due to relatively low concentrations of ozone precursors such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) occur in weekends. There are different hypotheses about weekend effect in the literature [11, 31, 33].

At all sampling sites, weekday and weekend SO_2 concentrations were quite similar. Since SO_2 levels are mostly associated with heating activities and unless heating activities are not changed significantly between weekday and weekend periods, there will not be important variabilities in SO_2 concentrations between weekday and weekends. Also, in Table 1, it can be seen that change in traffic density between weekday and weekend periods did not have significant effect on SO_2 concentrations.

3.3. Vertical Variations of the Pollutant Concentrations

There is limited data describing vertical profiles of the pollutants near roadways, particularly in urban areas. In urban areas there are not only variable concentrations of the pollutants at different sites but there is also a vertical variation. In Figure 7, 8 and 9, vertical variations of NO_2, SO_2 and ozone concentrations at Sampling Site 1 and 2 were given respectively.

As seen in Figure 7, NO_2 concentrations decreased with the elevation. Higher concentrations at the lower floors may due to effect of traffic emissions. The vertical distribution of air pollution may also vary depending on meteorological parameters and on the structure of the city [34]. At Sampling Site 1, at the highest floor (31. meter), NO_2 concentrations increased slightly. The reason for this case may be chimney gas emissions of the building since this floor was the last floor or smoking activities of the people in the balconies living in this floor.

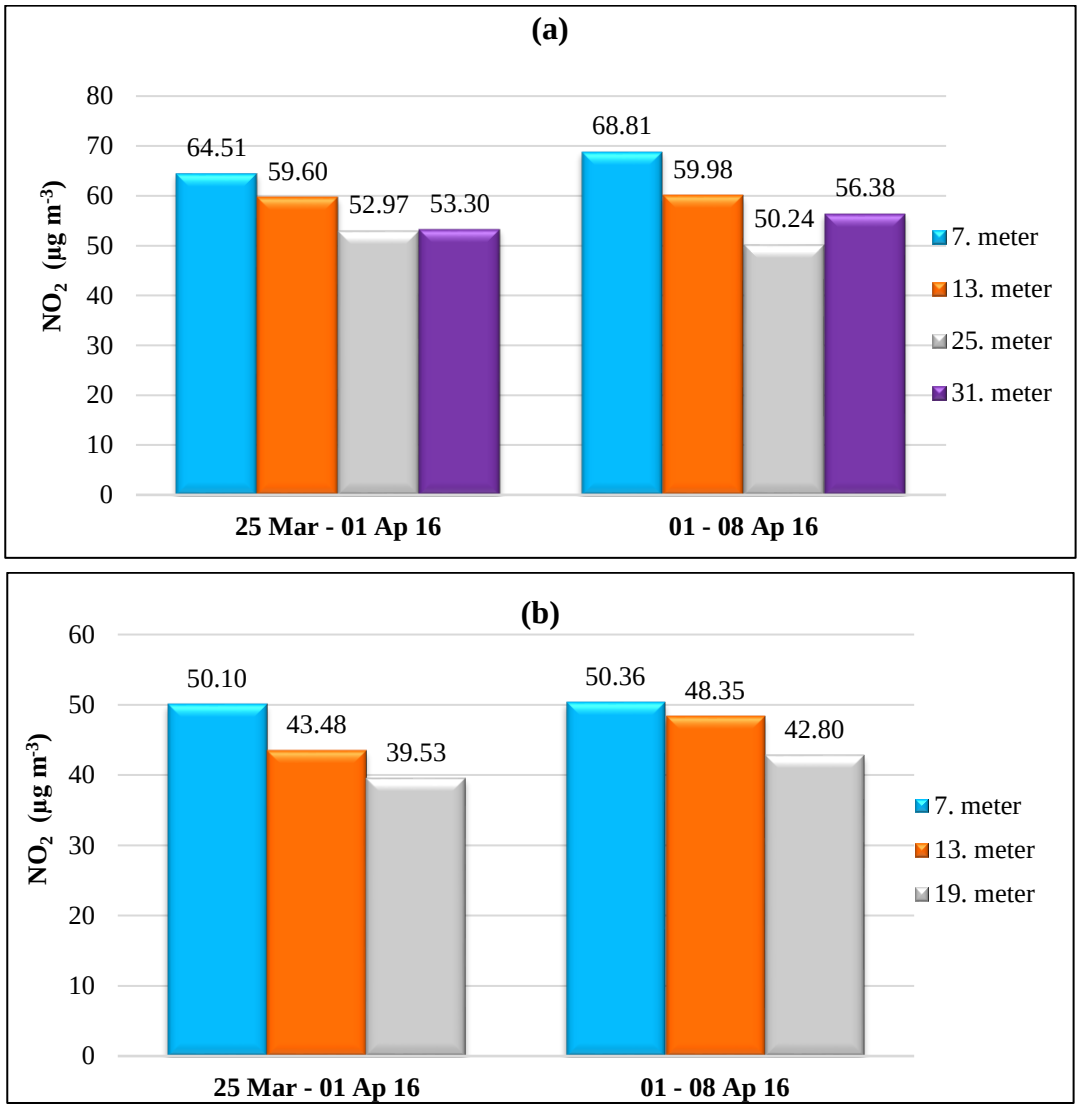

Figure 7. Vertical variations of NO_2 concentrations at (a) Sampling Site 1 and (b) Sampling Site 2

In Figure 8, SO_2 concentrations increased with the elevation slightly due to chimney gas emissions and this is an expected situation [35]. In urban areas, the main source for SO_2 concentrations is heating activities. Both coal and natural gas is used at Sampling Site 1 and 2 for domestic heating. So, highest concentrations were measured at the floors closest to the roofs of the buildings and so to the chimneys. Also, other reason for the increase of SO_2 concentrations with elevation may be transport of SO_2 rich air from the slum areas of the city where only low quality coal is used for heating.

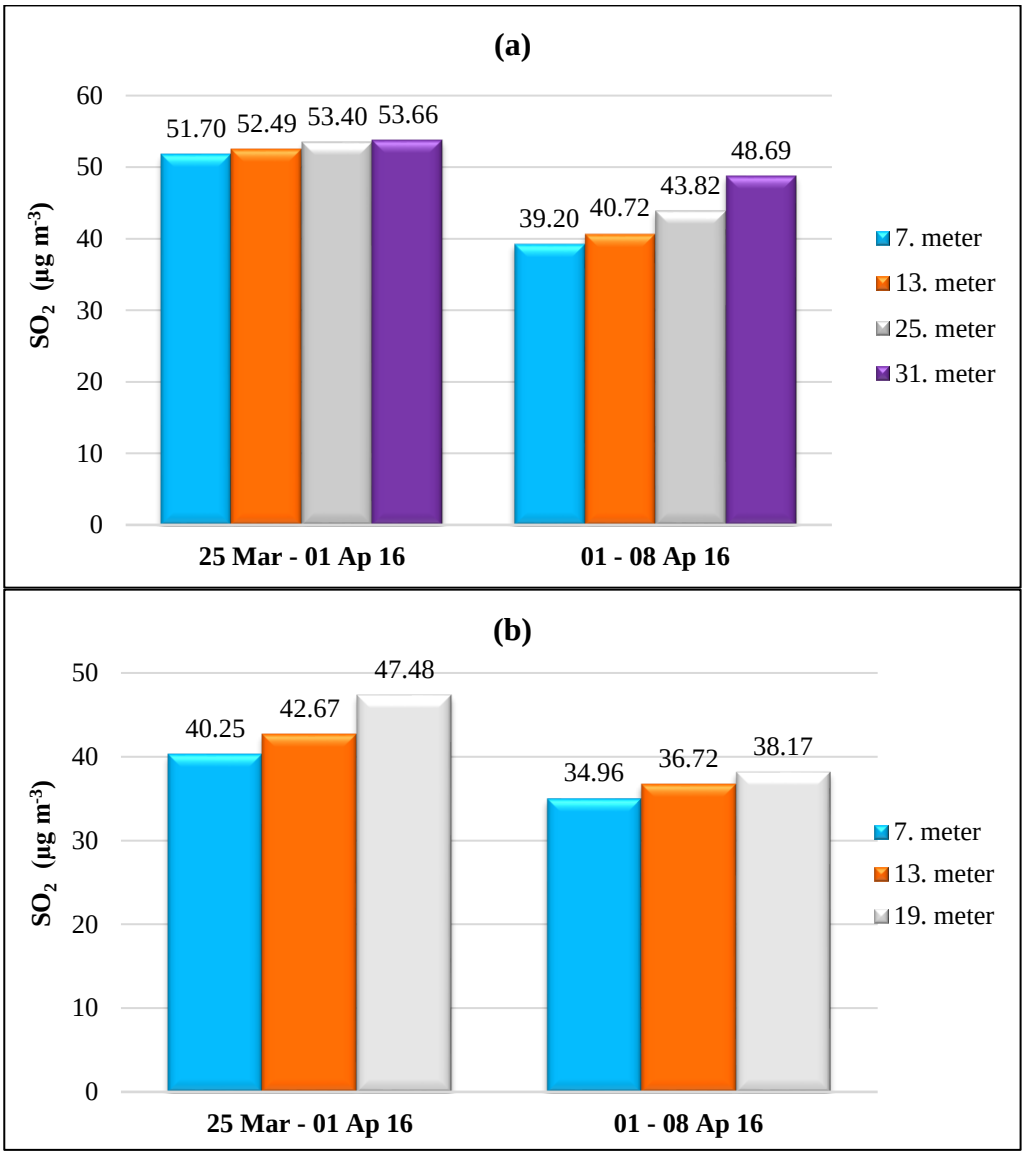

Figure 8. Vertical variations of SO_2 concentrations at (a) Sampling Site 1 and (b) Sampling Site 2

It has been known that ozone concentrations increase with elevation in the lower troposphere [36, 37]. Although the maximum sampling height in the study was not too much, an increase in ozone concentrations was observed with elevation from ground level to 25 m. The reason of lower concentrations near the surface may be surface deposition and also titration by some species emitted from the surface. Vertical variation of ozone concentrations was reverse case of NO_2 concentration variation. As NO_2 concentrations decreased with elevation, ozone levels increased at the same elevation range due to reduced destruction of ozone with decreasing NO_2 concentration with elevation. At Sampling Site 1, decreasing of ozone concentrations at 31 meter (Figure 9a) was associated with slight increasing of NO_2 concentrations at the same elevation (Figure 7a). Vertically reverse relationship between ozone and NO_2 concentrations was shown in Figure 10 for Sampling Site 1. Similar trend was also observed for Sampling Site 2.

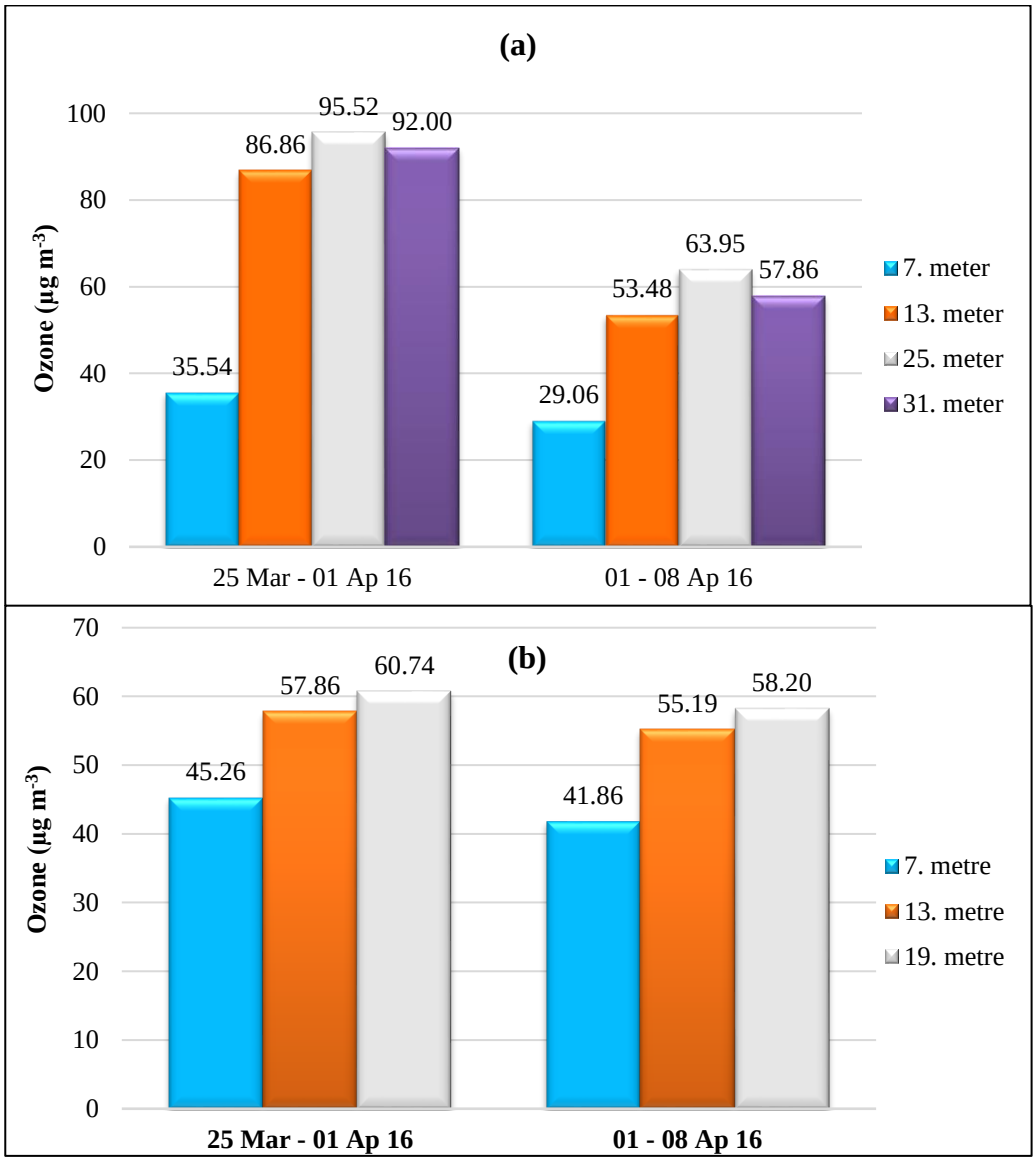

Figure 9. Vertical variations of ozone concentrations at (a) Sampling Site 1 and (b) Sampling Site 2

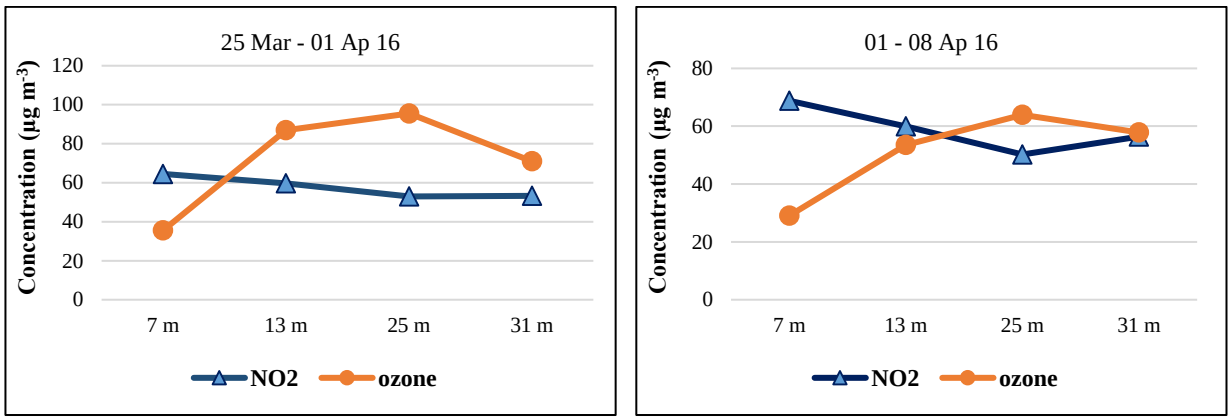

Figure 10. Reverse trend in vertical variations of NO_2 and ozone concentrations at Sampling Site 1

4. CONCLUSIONS

Ambient concentrations of NO_2, SO_2 and ozone were measured at three different sampling sites in the urban atmosphere of Eskişehir. The highest NO_2 concentrations were measured at the sampling site which is located in the city center and having urban, traffic and residential characteristics. Contrary to NO_2, higher ozone concentrations were measured at the sampling site located far from the city center. Spatial variations of SO_2 concentrations were similar to those of NO_2.

Weekday and weekend concentrations of the pollutants were measured separately and compared with each other. Weekday NO_2 concentrations were higher than weekend concentrations while higher ozone concentrations were observed in weekends. This result showed expected "weekend effect" for ozone. Weekday and weekend SO_2 concentrations were quite similar at all sampling sites.

Beside spatial variations, vertical variations of each pollutant were also investigated. NO_2 concentrations decreased with the elevation and reverse case was observed for ozone with increasing concentrations at the same elevation range. SO_2 concentrations increased with the elevation due to the effect of chimney gas emissions.

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