INVESTIGATION of TEMPORAL VARIATIONS in PARTICLE NUMBER CONCENTRATIONS of AMBIENT AIR in ANKARA and THE RELATIONSHIP BETWEEN PARTICLE NUMBER and MASS CONCENTRATIONS

by

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To my family

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ABSTRACT

INVESTIGATION of TEMPORAL VARIATIONS in PARTICLE NUMBER CONCENTRATIONS of AMBIENT AIR in ANKARA and THE RELATIONSHIP BETWEEN PARTICLE NUMBER and MASS CONCENTRATIONS

In this study, temporal variations in the number and mass distribution of atmospheric particles are investigated. Particle number distribution of particles was measured using a laser spectrometer at every minute for 12 months. In addition to particle number measurements, PM2.5 and PM10 samples were collected at the same time and at the same location. Particle mass concentrations were measured in the collected samples. Statistical tests are used to determine factors affecting variations in particle number concentrations. The effect of meteorological variations and the effect of anthropogenic and natural events on particle count and mass concentration data are analyzed.

In the study, a mathematical model is developed, based on the collected data, to convert the particle number concentration data to particle mass concentration data and evaluate the direct effect of meteorological data on the particle count concentration data. The model is verified by a detailed comparison of the results obtained from the model with the GRIMM mass concentration results.

ÖZET

ANKARA DIŞ HAVASINDA PARTİKÜL SAYISI VE PARTİKÜL AĞIRLIK ANALİZİ VE PARTİKÜL SAYISI İLE PARTİKÜL AĞIRLIĞI ARASINDAKİ İLİŞKİNİN İNCELENMESİ

Çalışmanın temel amacı, Ankara'da partiküler madde sayısal dağılımının zaman içerisindeki değişimini ve bu değişime katkıda bulunan faktörlerin ortaya çıkartılmasıdır. Bu amaçla Ankara atmosferinde bulunan partiküler maddenin sayısal konsantrasyonu ve boyut dağılımı, 12 ay boyunca, ODTÜ kamusunda, dakikalık olarak ölçülmüştür, Sayısal konsantrasyon ölçümlerinin yanında aynı zaman aralığında ve aynı noktada PM2.5 ve PM10 örnekleri toplanmıştır. Bu örneklerde partiküler maddenin kütlesel konsantrasyonu belirlenmiştir. Daha sonra elde edilen veri setine istatistik teknikler uygulanarak Ankara'da partiküllerin sayısal konsantrasyonların etki eden faktörler ortaya çıkartılmaya çalışılacaktır.

Dakikalık partikül sayısal değerleri, partikül kütle değerleri ve meterolojik faktörlerin etkisi de değerlendirilerek çoklu regresyon analiziyle sayısal partikül değerlerinden kütlesel değerlerin hesaplanmasını sağlayan matematiksel model geliştirilmiştir. Model sonuçları sayısal partikül ölçümü yapan cihaz tarafından belirli bir zaman için sağlanan kütlesel değerlerle karşılaştırılarak doğrulaması sağlanmaya çalışılmıştır.

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LIST OF SYMBOLS/ABBREVIATIONS

Symbol	Explanation	Unit
°C	Celsius	-
m^3	Cubic Meter	-
μm	Micrometer	-

Abbreviation	Explanation
WHO	World Health Organization
PM	Particulate Matter
EPA	Environmental Protection Agency
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
NOAA	National Oceanic and Atmospheric Administration

1. INTRODUCTION

Air Pollution is defined as the contamination of the indoor or outdoor environment by any physical, chemical, or biological agents (WHO, 2020). Not only in low-, middle-income countries but also in high-income countries are affected by outdoor air pollution which is a major environmental health problem.

Exposure to small particles such as PM10 and PM2.5 can affect both lungs and heart of human beings (Yin et al., 2017). Many studies showed the relation of exposure to particle pollution to health such as:

- premature death in people with heart or lung disease
- heart attacks that are non-fatal
- irregularities in the heartbeat
- aggravated asthma
- decreased lung function
- increased respiratory symptoms, such as irritation of the airways, coughing, or difficulty breathing (EPA, 2020).
- migraine headaches (Chiu et. al., 2015)

In a report prepared by WHO, 2016, it is found that air pollution is the reason for 4.2 million premature deaths worldwide per year. The reason for this mortality rate is the exposure of human beings to small particles which are defined as 2.5 micrometers or less (PM2.5) and 10 micrometers or less (PM10). These particles are found to be the main pollutants that lead to cardiovascular and respiratory diseases and cancers.

Studies show that particulate matter number concentration has a significant effect on both human health and the environment (Malley C., 2020) which leads us to work on this subject. In many of the studies (Gauderman et al., 2004, Pope et. al., 2002), exposure to particulate matter which has less than 10 μm aerodynamic diameter (PM10) or particulate matter which has less than 2.5 μm aerodynamic diameter (PM2.5) was analyzed. However, particle number concentration data might be more closely correlated with the health effect of particulate matter on human health than the mass concentration (Wichmann et. al., 2000). If particle number and mass concentration are determined at the same time particle number concentration data can give important information about the air quality

and also analyze the health effects working with particle count concentration data with mass concentration data will be beneficial (Lorelei A., 2020).

There are many advantages of working with a particle counter; easy to use, the cost is low and able to measure particle concentration for short time intervals. That is why particle counters are effective for determining spatial and temporal variations of particle concentrations (Tittarelli A et. al., 2008).

Since most of the studies are based on mass concentration data that are collected with mass measuring instruments, determining the relationship between particle count and mass concentration data is required.

The main purpose of this study is to determine the relationship between particle number and particle mass concentration and the temporal variation of particle number and mass concentrations in Ankara, Turkey, and the factors that are effective in this concentration. Effect of meteorology like temperature, wind, and rain on particle number concentrations will be discussed. Also, the contribution of the dust and anthropogenic events to the particulate matter concentration are discussed.

2. LITERATURE REVIEW

2.1. Sources of Particulate Matter (PM)

Major pollutants that affect human health and environment are carbon monoxide, ozone, nitrogen dioxide, sulfur dioxide and one of the most important ones is particulate matter which we will mostly focus on this thesis (Fierro, 2000, WHO, 2020). Particulate matter (PM) is the generic term that is used for a mixture of solid particles and liquid droplets suspended in the ambient air having physical and chemical diversity over a wide size range (Fierro, 2000, U.S. EPA, 2020, Hassan H. et. al., 2020). Particulate matter (PM) is an important and complex topic. Because it is one of the main pollutants for the environment, by scattering and absorbing light, and with that effect, particulate matter can change global and regional climate (Charlson et al., 1992; Kim et al., 2006, Malley et al., 2020), also particulate matter behaves as cloud condensation nuclei which affect cloud amount and lifetime are changed (Ramanathan et al., 2001; Sekiguchi et al., 2003, Xue et al.,2020). Particulate matter is also important for its health effect. Particles penetrate through lungs and with long term exposure to the particles, respiratory and cardiovascular health problems are occurred. (Pope et al., 2002, WHO, 2005). Particle deposition in the human respiratory system determines the hazard of particles to human health. Particle size, shape, and density affect deposition rates. But the most important characteristics are the size and aerodynamic properties (Fierro, 2000). Since particulates in the air don't have a spherical shape, it is not possible to calculate the size of the particulate matter. That is why a new term aerodynamic diameter is derived. The diameter of a spherical particle having a density of 1 mg/cm^3 that has the same inertial properties such as terminal settling velocity in the gas as the particle of interest is defined as the aerodynamic diameter (EPA, 2020).

Particulate matter can be emitted from natural and anthropogenic sources. In figure 2.1. (Whitby, 1978), a summary of the main natural and anthropogenic sources of particulate matter can be seen as primary and secondary PM. As can be seen, sources of particulate matter change for particles with a diameter higher and lower than 2.5 μm. All the particles are generated by natural or anthropogenic means. Main anthropogenic sources are road transport, stationary and non-stationary combustion processes, and other sources like forest fires and agriculture. Road transport is regarded as the dominant source of particulate matter in the urban areas since it is the direct emission from vehicle exhausts. Motor vehicles are one of the major sources of Nitrogen oxides are emitted in urban environments. Also moving vehicles results in resuspension of the road dust and fugitive dust from

paved and unpaved roads are produced. Stationary combustion sources are fossil fuel combustion for electrical utilities, residential space heating, and industrial processes. Coal-burning for residential space heating is the main source of particles in cities but with the decreasing usage of coal as the fuel, the problem is partially solved. More coal is used in power plants compared with residential usage. Even though filters are used to reduce particulate emissions, smaller fractions will be discharged. With the high stacks of power plants and wind, particles may reach the free troposphere and may be transported over regions. It is found that SO_2 is mainly produced as a result of the power plant working process. Industrial particulate emissions can be an important factor in urban areas. This factor depends on the source used, the process, and the precautions are taken. Construction, mining, cement plants, and ceramic factories are the main contributors to the ambient air particulate matter as non-combustion sources. Forest and agricultural fires are important factors since particles are emitted from fires and resuspension from burnt soils are major sources of particulate matter in the areas where there are constant forest fires and vegetation fires to create construction land and to clear area for agriculture. (U.S. EPA, 2020, TWG, 1997, Doğan, 2005) The main natural sources as can be seen from figure 2.1 are; sea spray, soil resuspension, volcanic emissions, and biogenic sources. Oceanic aerosols are one of the naturally produced particles with the action of wind on the ocean surface (Blanchard et al., 1980). Bursting of air bubbles on the sea surface and the breaking waves on the sea cause many droplets to be ejected from the sea. These droplets dry and leaves salt particles as suspended particles in the ambient air. Since the majority fraction of the salt particles has high diameters, most of the particles emitted from sea surface will sediment quickly and as a result sea salt will be most effective in coastal areas. But it is seen that a small fraction of particles is transported to the higher parts of the boundary layer and also into the free troposphere (Mats et al., 1999). Another natural source is soil resuspension which is one of the primary natural aerosols in the atmosphere. Soil dust is produced by a rock or sand weathering with different meteorological mechanisms such as wind, water or erosion. The composition of these dust particles may show differences with different regions depending on the geological source. Volcanic emissions are another important natural source that is composed of mineral fragments, fine grained rock. and glass shards, with acids, salts, and adsorbed gases coating particulate surfaces (Ruggieri et al., 2012). With all these compositions, particles emitted from volcanos is accepted as an important local source of ambient air pollution. Also, volcanos are one of the major sources of sulfur in the atmosphere with the high composition of sulfur dioxide (SO_2) and hydrogen sulfide (H_2S) (Aiuppa et al, 2005). Biogenic sources are important sources of natural particulate matter. Secondary particles produced from biogenic precursors make up an important fraction of the total PM (Wagener et al., 2012). The oxidation of a fraction of terpenes emitted by vegetation and reduced sulfur species from anaerobic environments leads to secondary

PM formation. (U.S. EPA, 2004). Also, it is found that Plankton activity in the oceans is an important source of secondary particle formation through the production of Dimethyl Sulfide (Ruggieri, et al., 2012).

	Sources					
	Primary (PM < 2.5 μm)		Primary (PM > 2.5 μm)		Secondary PM Precursors (PM < 2.5 μm)	
Aerosol species	Natural	Anthropogenic	Natural	Anthropogenic	Natural	Anthropogenic
Sulfate (SO_4^{2-})	Sea spray	Fossil fuel combustion	Sea spray	—	Oxidation of reduced sulfur gases emitted by the oceans and wetlands and SO_2 and H_2S emitted by volcanism and forest fires	Oxidation of SO_2 emitted from fossil fuel combustion
Nitrate (NO_3^-)	—	—	—	—	Oxidation of NO_x produced by soils, forest fires, and lighting	Oxidation of NO_x emitted from fossil fuel combustion and in motor vehicle exhaust
Minerals	Erosion and reentrainment	Fugitive dust from paved and unpaved roads, agriculture, forestry, construction, and demolition	Erosion and reentrainment	Fugitive dust, paved and unpaved road dust, agriculture, forestry, construction, and demolition	—	—
Ammonium (NH_4^+)	—	—	—	—	Emissions of NH_3 from wild animals, and undisturbed soil	Emissions of NH_3 from animal husbandry, sewage, and fertilized land
Organic carbon (OC)	Wildfires	Prescribed burning, wood burning, motor vehicle exhaust, and cooking	Soil humic matter	Tire and asphalt wear and paved road dust	Oxidation of hydrocarbons emitted by vegetation (terpenes, waxes) and wild fires	Oxidation of hydrocarbons emitted by motor vehicles, prescribed burning, and wood burning
Elemental carbon (EC)	Wildfires	Motor vehicle exhaust, wood burning, and cooking	—	Tire and asphalt wear and paved road dust	—	—
Metals	Volcanic activity	Fossil fuel combustion, smelting, and brake wear	Erosion, reentrainment, and organic debris	—	—	—
Bioaerosols	Viruses and bacteria	—	Plant and insect fragments, pollen, fungal spores, and bacterial agglomerates	—	—	—

Figure 2.1. Anthropogenic and natural sources of primary and secondary particles (Whitby, 1978).

2.2. Formation of Particulate Matter

Particulate matter can be found both in the troposphere and stratosphere. Properties of particulate matter change with the changing morphologic, chemical, physical, and thermodynamic conditions. Classification on the formation mechanism of particulate matter can be done in two parts named primary and secondary particles. Primary particles are directly emitted into the ambient air and the former one is as secondary particles which are formed in the atmosphere by the transport of a variety of emissions such as VOCs (Volatile organic compounds) and SOx (Sulfur oxides). Because of the complex composition of the PM, physical and chemical properties can be different and change with time, region, and meteorology (U.S. EPA, 2020). Particles with different sizes come from different sources as discussed previously.

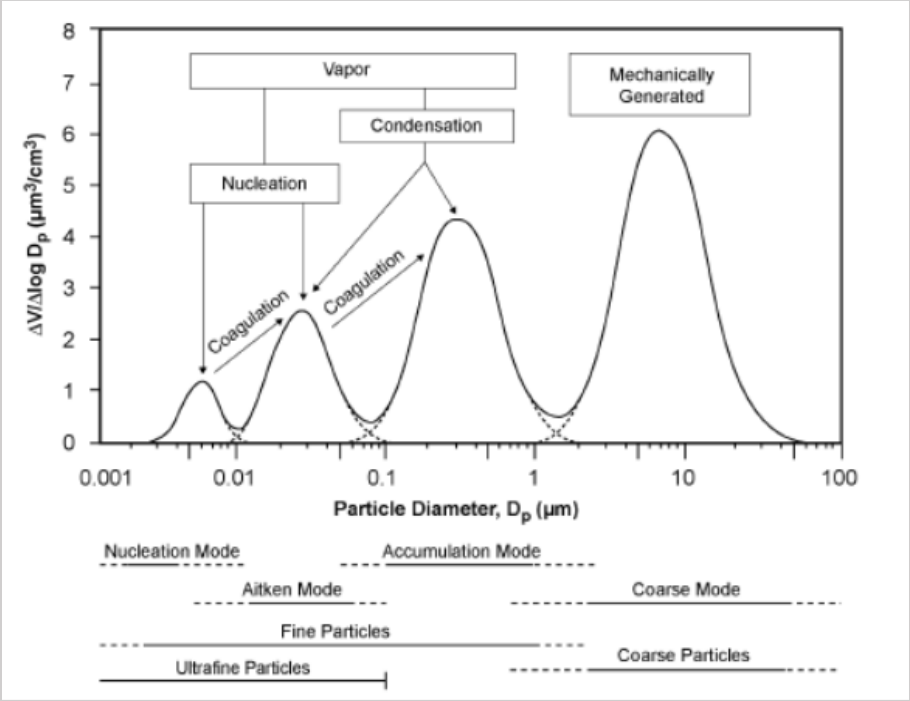

Figure 2.2. Contribution of particle mass to the volume. Whitby (1978).

In Figure 2.2, particle diameter vs. volume change with particle diameter is given. It is seen that much of the volume is composed of coarse and accumulation mode materials. Nucleation mode particles are freshly formed particles having an aerodynamic diameter of less than 10nm. They are formed as a result of the chemical compounds, such as ammonia and sulphuric acid, gas to particle conversion. Also, gasoline engines produce nucleation mode particles. Aitken mode particles are larger particles having a diameter between 10-100 nm. Aitken mode particles are mainly formed as a result of the nucleation or growth of smaller particles, traffic emissions, and diesel engines. Accumulation mode particles have an aerodynamic diameter between 0.1- 1 μm. The main sources

of accumulation mode particles are industrial combustion, natural sources like a bubble burst over the ocean. Coarse mode particles are the particles having a diameter higher than 1 μm. These particles are generally produced as a result of mechanical processes, industrial processes, resuspension (Laakso et al., 2003, U.S. EPA, 2004).

2.3. Properties of Particulate Matter

PM10 and PM2.5 are the particulate matter with an aerodynamic diameter of less than 10 μm and 2.5 μm, respectively. The PM10 fraction is defined as inhalable coarse particles since they can be deposited in the upper respiratory system and the PM2.5 fraction is called fine particles (Fierro, 2000, U.S. EPA, 2010).

2.3.1. Size distribution

In order to determine properties, effects, and fate of particulate matter, proper determination of particle size is an important parameter. Residence times of particles and as a result deposition in the atmosphere, deposition of particles in the lung, and light scattering are directly connected with the size of the particulate matter. As a result, the size distribution of particles is a major factor affecting visibility, radiative balance, and climate. By using cascade impactors or cyclones, particle size distribution is measured in the range from 1 nm to 100 μm and by determining the size distribution of particulate matter, detailed information on mode distributions and information about the formation and transformation of particulate matter can be understood (U.S. EPA, 2010, Whitby, 1978).

2.3.1.1. Number Distribution of Particulate Matter. In general, ill-health is related to particles in the atmosphere is due to the mass concentration of particulate matter. Different studies (EPA, 2020, Chiu et. al., 2015) showed that exposure to fine particles may lead to health problems. Small particles such as PM 2.5 and PM 1 can reach deep into the lungs, and much smaller particles (<100 nm) may pass through the lungs to affect other organs and they cause environmental pollution. But if the mass concentration is used to determine fine particle concentration, the result will not be accurate because these particles do not contribute much to total mass concentration even when they dominate the particle number concentration (Oberdorster 2001; Branis et al. 2005, McCawley et al., 2001).

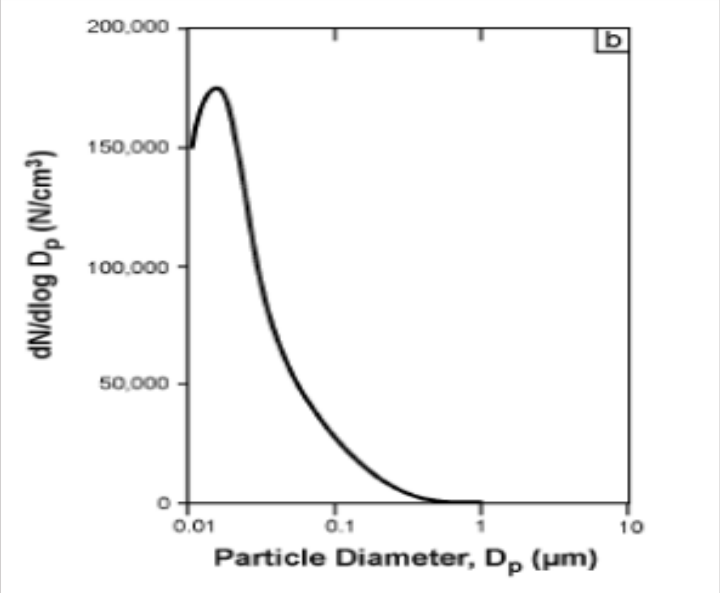

Figure 2.3. Change of particle diameter for particle number concentration (Whitby, 1978).

Whitby (1978) showed the relation between number concentration change with particle diameter in figure 2.3 and it is seen that most of the particles are quite small (smaller than 0.1 μm) but as discussed before in figure 2.2, coarse and accumulation mode particles with an aerodynamic diameter higher than 0.1 μm contribute most of the particle volume (U.S.EPA, 2004).

Also, particles with a diameter between 0.1 μm-1 μm (accumulation mode particles) have an important effect on the radiation field in the atmosphere by scattering light and acting as cloud condensation nuclei. (Matsumoto et al., 2008)

In many studies, it is seen that effects of small particles are important in many ways (like health, light scattering, etc.) so because of the importance of these particles with an aerodynamic diameter less or equal to 10 μm (PM10), 2.5 μm (PM2.5), and 1.0 μm (PM1.0) number concentration measurement researches are applied more in many industrial and working places in the last years. (Halek, 2009)

2.3.1.2. Surface area, volume, and mass distribution. Using mass concentration for the analysis of the toxicity of ultrafine particles may not give correct results since particles with high mass does not contribute much to the number concentration. As a result, an alternative method, surface area distribution is used. This distribution is biologically more relevant and can give more relevant results than other exposure metrics because surface reactivity may be the reason for adverse health effects of ultrafine particles and most of the part that contributes to surface area distribution is not much

effective in the mass concentration. Many studies showed that regardless of the particle composition, size, and shape, health problems in the lung caused by ultrafine and fine particles are proportional to surface area deposited. (Park et al., 2008)

In research done by analyzing more than 1000 size distributions in the United States, volume, surface area, and number distributions are plotted and given in figure 2.4. In figure 2.4. three different graphs are compared that are particle number concentration change with particle diameter, surface area change with particle diameter, and volume change with particle diameter. Distributions show that most particles are small (mostly below 0.1 μm) but most of the particle volume and as a result particle mass is found for the particles with a diameter of more than 0.1 μm. (Whitby, 1978)

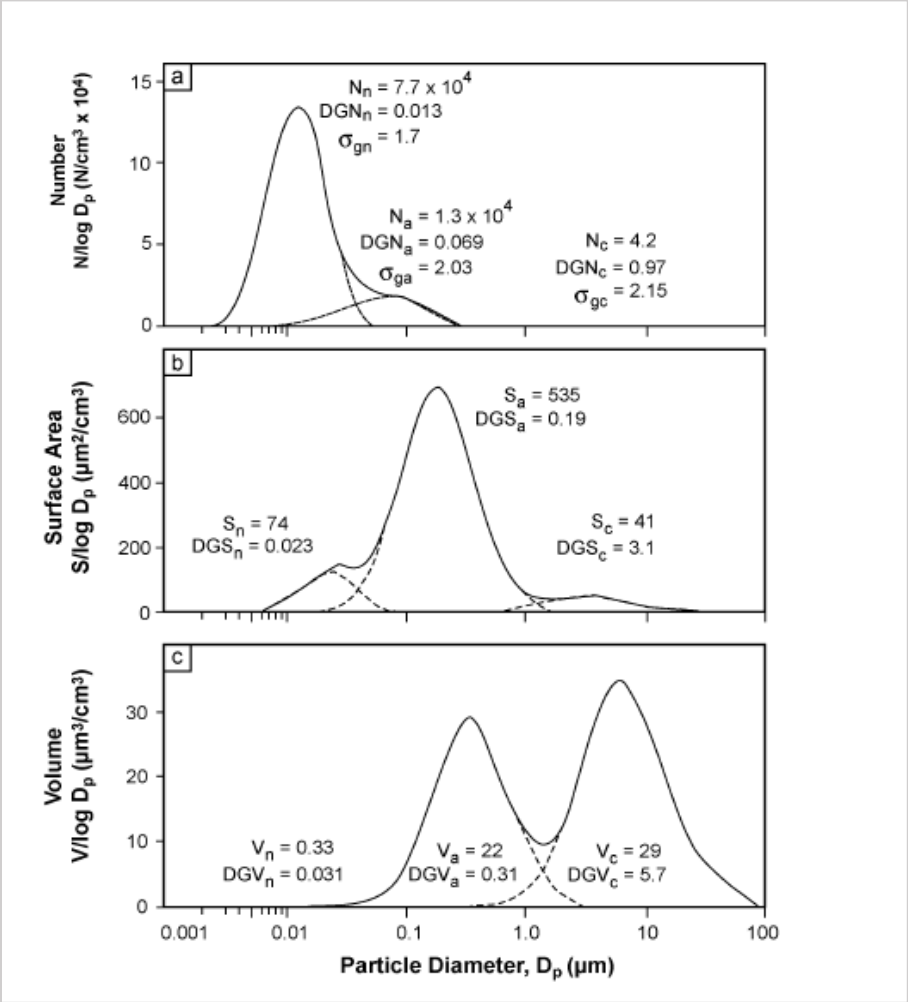

Figure 2.4. Distribution of coarse (c), accumulation (a) and nuclei (n) mode particles by three characteristics: (a) number, N; (b) surface area, S; and (C) volume, V. DGV= geometric mean diameter by volume; DGS= geometric mean diameter by surface area; DGN= geometric mean diameter by number; Dp= particle diameter (Whitby, 1978).

2.4. Removal of particulate matter from the atmosphere

The atmosphere is believed to be the major pathway of anthropogenic and natural contaminants, examples including organochlorines and some heavy metals found in tissues of arctic animals for anthropogenic sources and transportation of dust from Saharan desert to the surrounding areas such as Turkey, Greece, Iraq, and Saudi Arabia can be given (Pongkiatkul et al., 2007). Transport of these contaminants over exceeding national borders is a major air quality problem in urban areas. The travel distance of particles is determined by their residence time in the atmosphere. The residence time of the particles in the atmosphere is mostly dependent on the size of the particles.

Average residence time is calculated by determining the average lifetime of molecules. For particulate matter, the residence time is used to show how long a particle stays in the atmosphere before wet or dry deposition (Seinfeld, 2006).

Coarse particles have a short residence time because of its high mass. Also, Aitken mode particles have short residence time since they coagulate quickly and form bigger particles. These particles are quickly removed from the atmosphere. But accumulation mode particles have long residence time. The residence time of these particles depends on their solubility. If the particle is soluble, removal is done with precipitation having a residence time nearly 10 days equal to that of water. But if the particle is hydrophilic, residence time increases to at most 46 days for the particle to be wetted. Depending on the meteorological conditions, particles emitted can transport around the globe within a few days to a few weeks. For example, it is found that dust from the Saharan desert mostly found in the Mediterranean region but also some fraction is transported to Europe and America (Stefan et al., 2010, Pongkiatkul et al. 2007, TWG, 1997).

For the removal of trace gases and particles there are two ultimate paths:

1) Dry deposition
2) Wet deposition

2.4.1. Dry deposition

Dry deposition is defined as the transport of particles from the atmosphere onto surfaces when there is no precipitation. Affecting factors of the dry deposition can be given as; the level of atmospheric turbulence, the main properties of the deposited particle, and surface parameters. The turbulence level is effective mostly in close places to the ground level and determines the particle deposition rate down to the surface. Properties like size, density, the chemical composition may be the main determinants if the particle will be captured or not. Also, surface parameters are important since for example depending on the reactivity of the surface, deposition may not occur if the surface is nonreactive (U.S. EPA, 2004).

Main types of dry deposition:

Gravitational sedimentation: Removal of particles with the effect of gravity

Interception: Collision of particles to a close obstacle while following the streamlines.

Impaction: Impaction of small particles to a bigger obstacle which disables the particle to move in its curved streamline of the flow due to their inertia

Diffusion or Brownian motion: Random movement of aerosol particles due to collisions with gas molecules.

Turbulence: Turbulent eddies in the air transfer particles which can collide. (Seinfeld, 2006)

2.4.2. Wet Deposition

Wet deposition is defined as the removal of pollutants by cloud and fog drops, rain, snow, and delivered to the earth`s surface.

Main types of wet deposition:

Precipitation scavenging: Defined as the removal of particles with the raining cloud.

Cloud interception: This deposition occurs when cloud droplets impact on the terrain (Mostly at the top of the mountains)

Fog deposition: Defined as the removal of particles by settling fog droplets

Snow deposition: removal of the particle with snow

In order to get an effective wet deposition, three steps are necessary. First, condensed water must be present where gas and aerosol species are. Then previously defined wet deposition types should occur, and finally deposited form of the particle should reach the earth`s surface. In each of these steps, a particle can have chemical transformations (Seinfeld, 2006).

Accumulation mode particles (with a diameter of 0.1- 1 μm) mainly settled to the ground by cloud processes. If these particles consume hygroscopic components, particles will grow with the increasing relative humidity and they will serve as cloud condensation nuclei and finally if these particles get into cloud droplets. Particles will be removed with rain if cloud droplets grow enough to for rain. Coarse particles will be removed by impacting the falling raindrops. Ultrafine particles may diffuse into the falling raindrop and as a result, it will be removed from the air (U.S. EPA, 2004).

2.5. Effect of Particulate Matter

2.5.1. Health effects of particulate matter

During recent years, studies done to define the effect of particulate matter on health increased considerably which is a result of the correlation between particulate matter concentration, and mortality and hospitalization. (Ketzel et al., 2004) Because of this correlation, particulate matter mass concentrations (PM2.5 and PM10) are regulated in the US, EU, and TURKEY.

Annual and 24-hour limit concentrations used by EPA for PM2.5 are 12 $\mu g/m^3$ and 35 $\mu g/m^3$ respectively. For PM10, 24-hour limit concentration is used which is 150 $\mu g/m^3$. EU determined the annual PM2.5 limit concentration as 25 $\mu g/m^3$ and for PM10, 50 $\mu g/m^3$ limit is determined for the 24-hour period and 40 $\mu g/m^3$ for an annual period. In TURKEY as per Air Pollution Control Directive, 2009, there is no PM2.5 standard. The same limit concentration for PM10 in the EU is used in TURKEY.

Fine particulate pollution (PM2.5), because of its small size these particles can reache deeper in the lungs and also high concent of toxic metal and acids. (Fierro, 2000) As a result of many studies, the main health problems associated with particulate matter concentration are found as, premature death, aggravated asthma, acute respiratory symptoms, chronic bronchitis, decreased lung function, work, and school absences. Also, with a high surface reactivity of fresh soot particles, carcinogenic compounds may be carried with particulate matter (Highwood K., 2006, U.S. EPA, 2020). Mainly

elderly, children, and individuals with preexisting heart or lung disease are affected by particulate matter in the ambient air. It is estimated that tens of thousands of elder people die prematurely due to the exposure to fine particulates in the air. Also, many hospital admissions are associated with lung and heart diseases of elder people as a result of the particulate matter exposure. Children are affected by particulate matter since they breathe more than an adult and since respiratory systems of children are still developing, they are more susceptible to health problems caused by particulate matter. People with preexisting lung diseases are also adversely affected by particulate matter exposure. An increase in the hospitality rate, higher premature death is observed with increased exposure to fine particles. (U.S. EPA, 1997)

2.5.2. Effects of particulate matter on visibility

The particulate matter affects the visibility by scattering light in both urban and rural areas. Mainly all particles other than elemental carbon and some crustal minerals, scatter light and light scattering is the most effective light extinction component. It is found that large particles scatter more light having equal properties. The highest light scattering is observed for particles ~0.3-1.0 μm. (U.S.EPA, 2020)

Haze is one of visibility reduction situation happened when sunlight encounters tiny pollution particles in the air. Some light is scattered, and some are absorbed and as a result, visibility is reduced. More pollutant means more scattering and absorption of light which means increased visibility reduction. (Grantz et al., 2003)

Visibility reduction also directly affects people`s daily activities and enjoyment. It is found that people are emotionally affected by poor visibility and overall wellbeing is diminished. (U.S. EPA, 2009)

2.5.3. Effects of particulate matter on climate

Particulate matter has many effects on the climate. The direct effect is the light scattering of particulate matter which results to brighten the planet. Increased cloud brightness, changes in precipitation rate, and change in the cloud lifetime are the indirect effects of particulate matter on climate. As a result of the direct and indirect effects of particulate matter on climate, Earth`s albedo

and reflectance are affected which results in reduced sunlight reaching the surface. As a result, climate cooling is observed. (U.S. EPA, 2020).

2.5.4. Effects of particulate matter on ecology

The particulate matter affects ecology in many ways such as direct effects on the metabolic process of plant foliage, alteration in soil metal concentration, change in plant growth, animal growth, and reproduction. Since particulate matter is composed of a mixture of different originated particles, effects on the ecosystem are dependent on the chemical composition of the particulate matter (Grantz et al., 2003, U.S. EPA, 2020, U.S. EPA, 2004).

Measurable effects of particulate matter can be given as a change in photosynthesis efficiency, changes in soil salinity, and foliar effects. Particles settled or transferred to the foliar surfaces may be present for a long period on the leaves or bark surfaces which is taken by the plant through leaf surface or resuspended to ambient atmosphere by washing or wind. Any particulate matter settled on plant parts have mainly chemical effect depending on the composition of the particulate matter (U.S. EPA, 2020, Farmer, 1993).

2.5.5. Effects of particulate matter on materials

Building materials are affected by environmental elements such as wind, moisture, and sunlight. In order to protect the metal from corrosion, the oxidized film is applied. But with exposure to the anthropogenic pollutants, natural corrosion of metal is enhanced. Particulate matter also affects the painted surfaces and other building parts by soiling. As a result of the soiling, transmission of light is reduced in transparent materials, and reflectance of opaque materials is changed. In order to remove soiling, frequent cleaning or washing is required which decreases the usefulness of the material also depending on the surface repainting that may be done (U.S. EPA, 2009).

2.6. Plan of the Thesis

Chapter 1 introduces the problem and presents the aim, scope, and context of the thesis study.

In Chapter 2, the literature review for the subject was provided.

In Chapter 3 of the thesis, information about the sampling location, collection of samples with stacked filter units, measurement of particle number concentrations, and back trajectory calculations are described.

In Chapter 4, the relation of number concentration with mass concentration, meteorological factors, and dust events are given.

Chapter 5 concludes the study and the recommendations for future studies are presented in this chapter.

3. METHODOLOGY

In this chapter, information about the sampling location, collection of samples with different equipment, and trajectory calculations done are given.

3.1. Sampling location

A sampling of both particulate matter mass and number concentrations was done in Environmental Engineering department (Latitude: 39°53'12.07"N and in decimal degrees latitude: 39.886686 Longitude: 32°46'59.97"E and in decimal degrees longitude: 32.783325) of Middle East Technical University (METU), Ankara between August 2008 and January 2009. The location of the sampling site is given in figure 3.1.

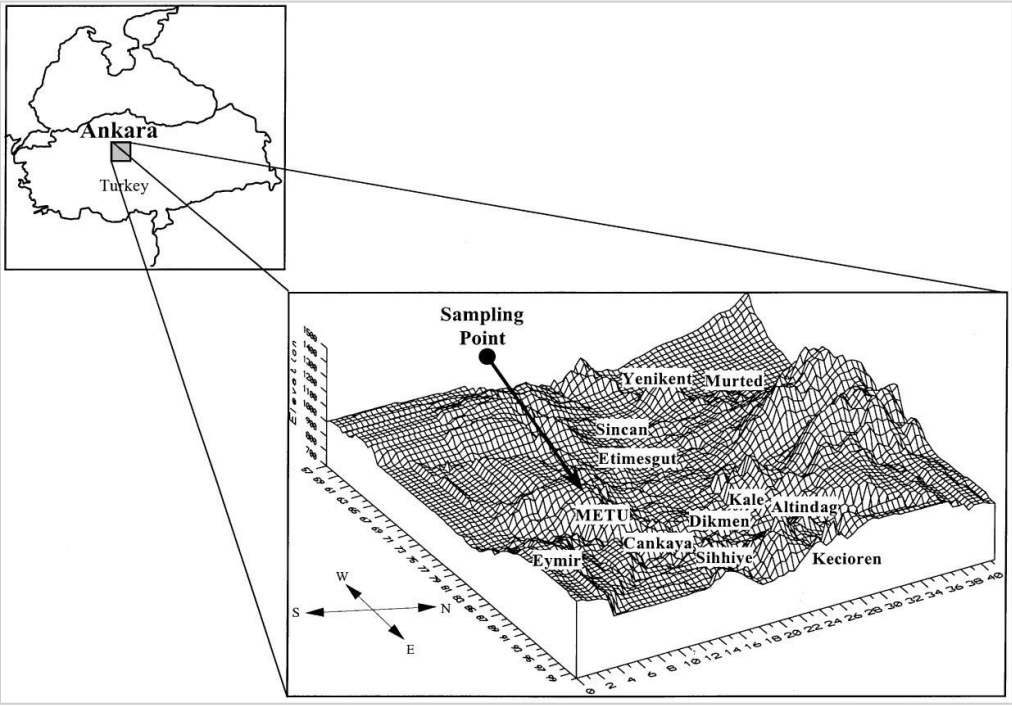

Figure 3.1. Sampling site location (Yatin et al., 2000).

Distance between the sampling station and the nearest road is nearly 50 m. Since the sampling station is stationed far corner of the center of METU, traffic is not heavy. Also, the distance of the sampling station to the nearest highway is nearly 2700m so the area can be defined as suburban. Source of pollutants such as traffic and combustions are not dense too much when compared with the center of Ankara so it can be considered that the samples show the background level of pollution in Ankara.

3.1.1. Meteorological Station

Meteorological data is taken from the Turkish State Meteorological Service. Since sampling location is a suburban area, Etimesgut station which is on the outskirts of the city is selected. The Etimesgut meteorological station is located on 39° 93` in latitude and 32° 65` in longitude. Surface and aloft meteorological data obtained from the Etimesgut station consist of hourly measurement of wind speed, wind direction, temperature, precipitation, and relative humidity and morning and afternoon mixing height values that are calculated from the radiosonde data.

3.2. Collection of samples with stacked filter units

Samples were collected using a stacked filter unit (SFU). In figure 3.2 general schematic of the sampling system can be seen. This unit has two nucleopore filters in series which are 8.0 and 0.4 μm respectively and a preimpactor to remove particles with diameters larger than 10 μm. The pump used to pull air has a 16 l/min flow rate which is essential for particle filtration, with this flow rate the unit should act as a dichotomous sampler (Hopke et al., 1997). In the literature, it is shown that with a pump operating at 16 l/min flux rate, particles with diameters larger than 2.2 μm (coarse fraction) is held in the top filter and smaller particles pass the coarse filter and held on the fine filter (Hopke et al., 1997, Yatin et al., 2000).

The sampler was installed on one side of the sampling station as given in figure 3.2 including the pump, flow control system, and rain shield. The collection of filters is done with 24-hour periods. Totally 88 coarse and fine samples are collected for the total 12 months sampling time.

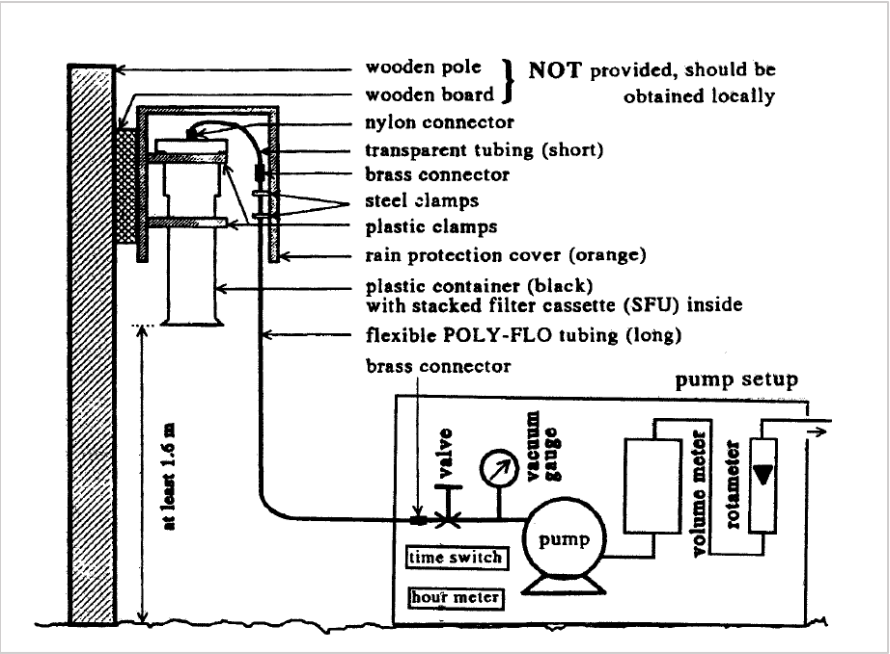

Figure 3.2. Parts of stack filter unit (Hopke et al, 1997).

3.3. Measurement of particle number concentrations

In order to count particles, GRIMM Ambient Aerosol Monitor with Integrated Sampling Pipe Heater Model 265 is used. To determine PM concentrations accurately, both volatile and non-volatile fractions should be measured. In filter sampling, volatile components may be lost because of the on-going sampling, to gas-solid, or even fluid-solid reactions. Some samplers heat the sampling probe to prevent condensation but heating volatile fraction causes less accurate results. The sampler that we used to determine particle number concentrations uses a different technique. An internal pump that is working at 1.2 L/min is used to collect samples. With this pump, sheath air is generated. In order to prevent dust, contamination is an optic device, this sheat air is used, it is used for the reference-zero tests during the self-test. The instrument first determines the concentration of volatile and non-volatile particles in the ambient air with the standard non-heated sample inlet. Then the sample inlet is heated to 80°C, with this temperature volatile fraction is removed and the non-volatile fraction of the sample is measured. Heated and non-heated measurements last 10 minutes separately.

The GRIMM instrument uses light-scattering technology to count particles, as a light source a semiconductor-laser is used. In figure 3.3, the general scheme of the light-scattering process is shown. As a single-particle pass through the laser, light scatters and this light is collected at approximately 90° by a mirror and sent to a recipient diode. A multi-channel size classifier is used to record the signal. The signal transmitted to the channels is classified by a pulse height analyzer.

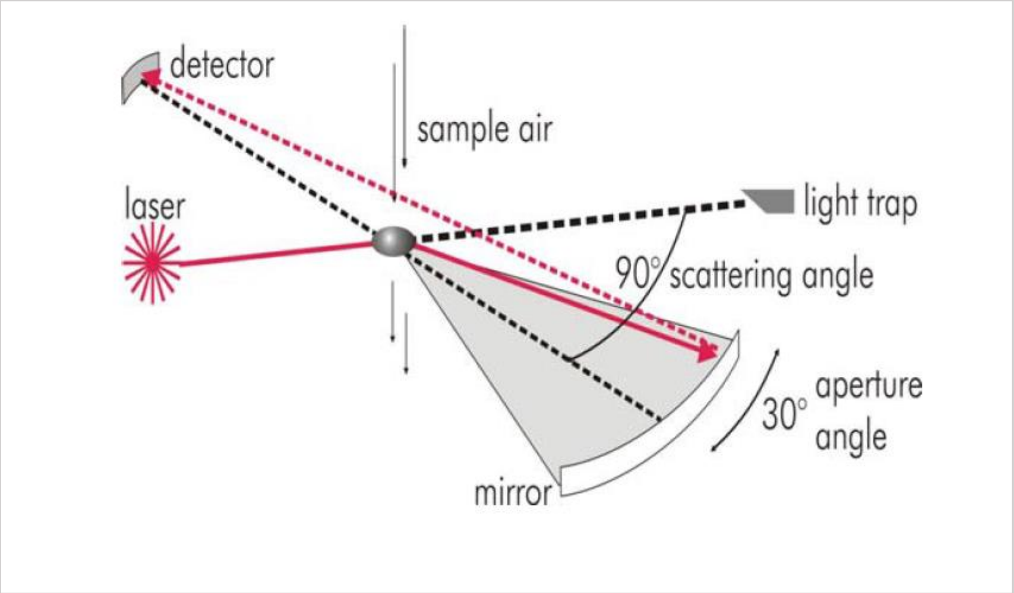

Figure 3.3. Particle counter light scattering process.

This system can measure the particles by their sizes in 32 bins, between 0.25 µm to 32 µm and particle mass between 1 to 1500 µm/m^3.

Data is collected for every minute in 12 months of the data collection period.

3.4. Back trajectory Calculations

Data for the back-trajectory calculations are obtained by using the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model from the NOAA website. HYSPLIT model by the National Oceanic and Atmospheric Administration (NOAA) at the Air Resources Laboratory (ARL). This model is a complete system used to compute trajectories of different dispersion models and deposition simulations by using different particle approaches. Many computer analyses and forecasts are done by the National Weather Service's National Centers for Environmental Prediction (NCEP). One of the operational systems is the GDAS (Global Data Assimilation System) which is the system used in this thesis. Data is collected every 12 hours for the 12 months data collection period. Coordinates of METU, Ankara (39.886686-32.783325) is used for the trajectory calculations. In the procedure of model calculation, three ground-level heights should be used. There is not a rule or an accepted height for the ground level height in the literature. Choices mainly depend on previous experience (Dogan et al., 2008). In a study done to understand long-range transport patterns in Thessaloniki, it is found that the 1500 m altitude can show the flow within the boundary layer

(Katragkou et al., 2009). In another study, it is found that 500 m altitudes can be used to determine long-range transport since this altitude ensures that the trajectory starts in the atmospheric boundary layer (ABL) (Karaca et al., 2009). By searching the previous studies and experience, five-day back trajectories for every 12 hours at arrival altitudes of 100m (red), 500 m (blue), 1500 m (green) shown in figure 3.4 in Metu (Middle East Technical University), Ankara were calculated for the 12 months data collection period using HYSPLIT. A total of 648 back trajectories were calculated.

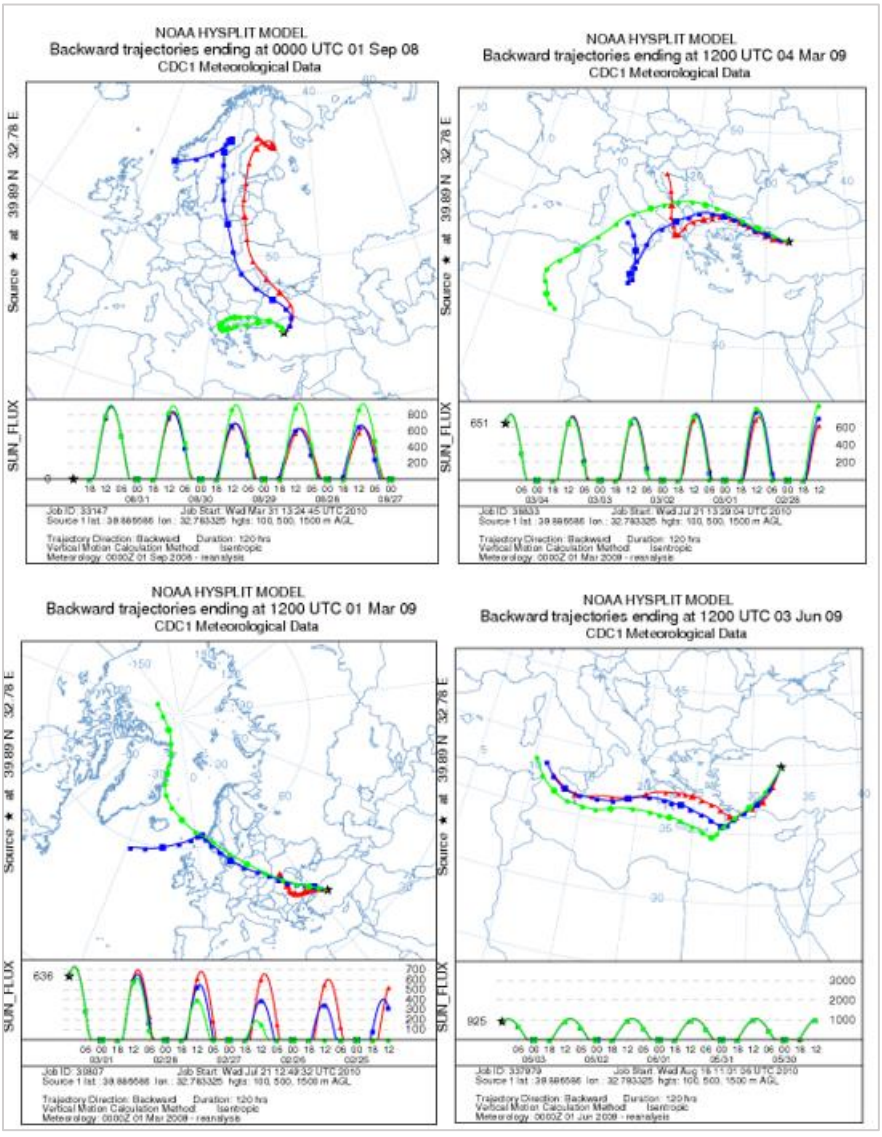

Figure 3.4. Sample Hysplit model results for dust transport, 100m (red), 500 m (blue), 1500 m (green).

3.5. Data Analysis Techniques

The concentrations of measured parameters are categorized by per minute, per hour, and per day for all particulate matter mass and number concentration data. After categorizing the data means, standard deviations, and medians of the data are constructed in order to show simple differences and similarities between pollutants and stations.

The relationship between air pollution and different meteorological factors is also investigated by simple regression analysis. Finally, multiple regressions are performed to construct a simple mathematical model to calculate particulate mass concentration from particulate number concentration.

4. RESULTS AND DISCUSSION

4.1. Data set

A study to see whether particle count concentrations show normal or log-normal distribution is given here.

Table 4.1. Sample data set and One-Sample Kolmogorov-Smirnov Test of particle count concentration data.

	Particulate matter particle count concentrations (count/L)			
	PM 0.25	PM 0.65	PM 1	PM 2.5
N	7550	7550	7550	7550
Normal Mean	191636.4131	2928.0557	786.1812	190.6684
Skewness	2.543	14.852	44.563	5.059
Std. Error of Skewness	0.028	0.028	0.028	0.028
Std. Deviation	1.83511E5	12530.68182	2283.05905	231.34074
Kolmogorov-Smirnov Z	15.394	35.530	31.880	17.850
Asymp. Sig. (2-tailed)	.000	.000	.000	.000

In table 4.1. since skewness values are positive and higher than the two times of standard error of skewness, it can be said that the data set is sloping to the right.

A significance level of 0.00 which is lower than 0.05 shows that mass concentration distribution is not normally distributed.

Table 4.2. Sample data set and One-Sample Kolmogorov-Smirnov Test of logarithmic particle count concentration data.

	Particulate matter particle count concentrations (count/L)			
	Log (PM 0.25)	Log (PM 0.65)	Log (PM 1.0)	Log (PM 2.5)
N	7550	7550	7550	7550
Normal Mean	191636.4131	2928.0557	786.1812	190.6684
Skewness	-0.236	0.495	-0.199	-0.631
Std. Error of Skewness	0.028	0.028	0.028	0.028
Std. Deviation	1.83511E5	12530.68182	2283.05905	231.34074
Kolmogorov-Smirnov Z	15.394	35.530	31.880	17.850
Asymp. Sig. (2-tailed)	.178	.124	.093	.074

In table 4.2. the logarithmic data set is prepared in order to see if the distribution is log-normal. Skewness values are found near 0 which shows distribution maybe log-normal. In figure 4.1. P-P plot is given as seen in the figure data set is log-normally distributed. Also, the One-Sample Kolmogorov-Smirnov Test is applied, table 4.2, to the logarithmic data set and as a result of the test Asymp. Sig. (2-tailed) the value which is also known as the p-value is higher than 0.05 and also in figure 4.2. lognormal P-P plot shows linearity, as a result, it can be said particle count data is log-normally distributed which is also parallel with the results of a similar study done by Lu, H.C. in 2003. Log-normal distributions of four selected random particle count concentration data are given in figure 4.2.

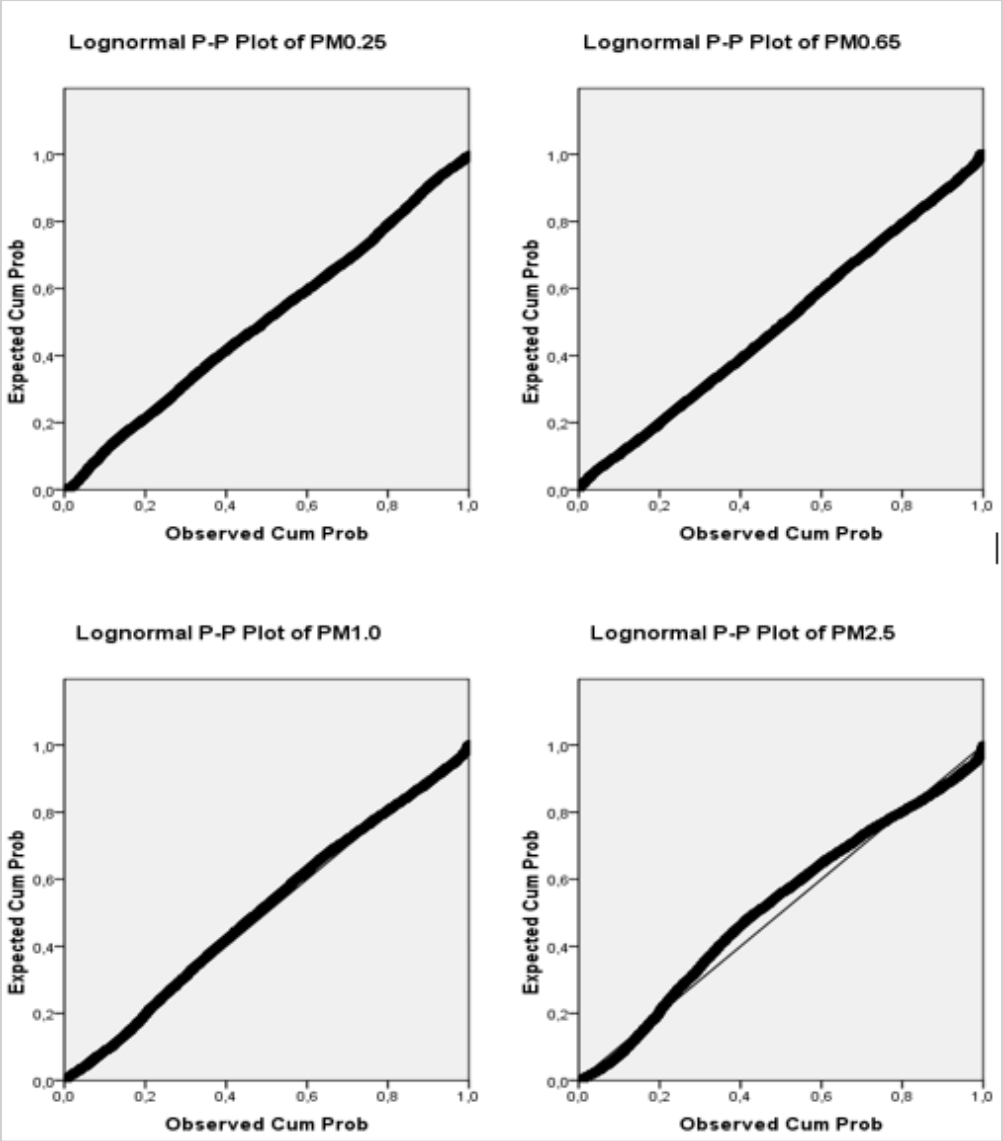

Figure 4.1. Lognormal P-P plots of different particle size.

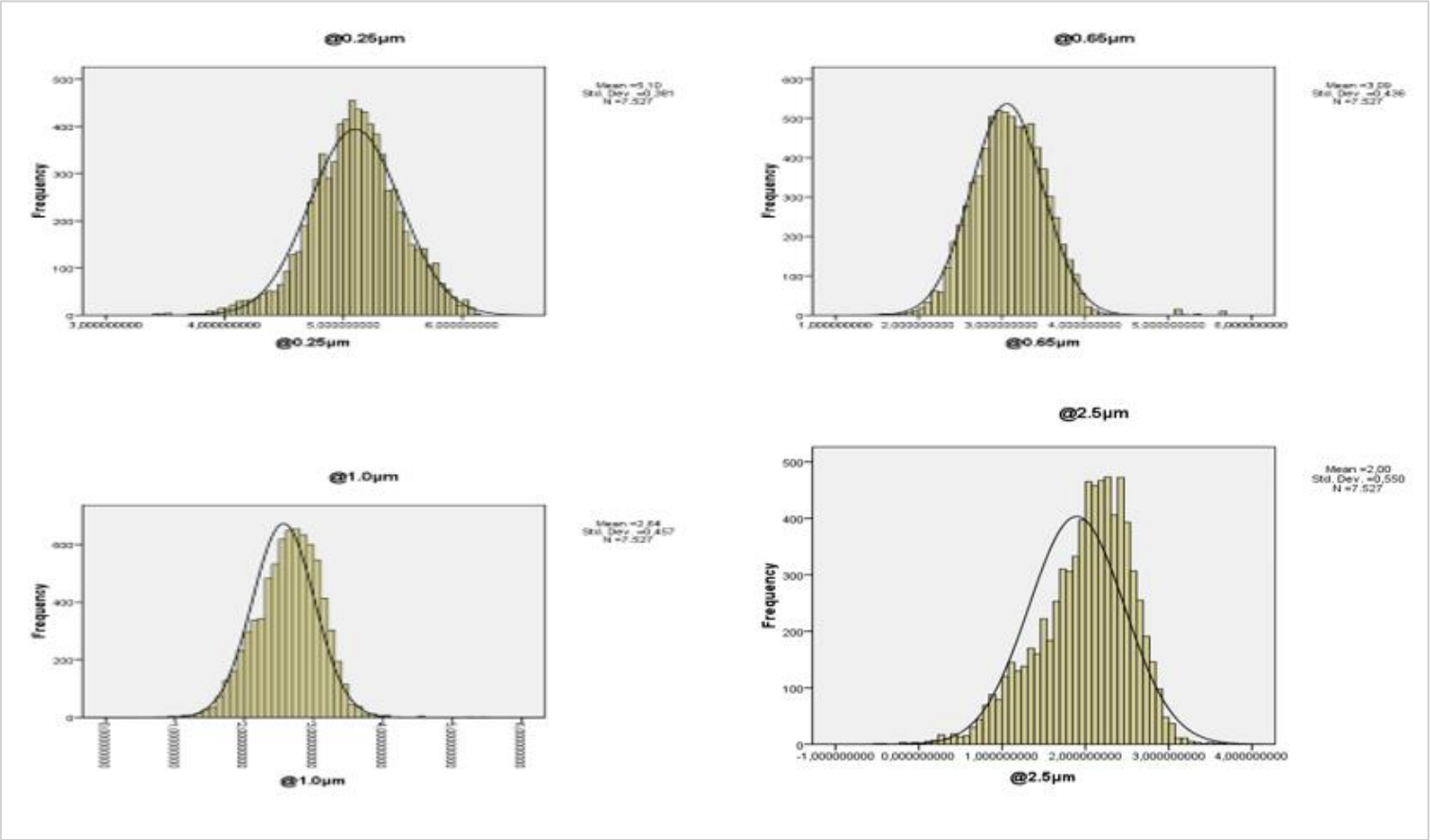

Figure 4.2. Log-normal distribution for particle count concentration of different sized particles.

PM 1, PM 2.5, and PM 10 mass concentration data are observed to see the distribution type.

Table 4.3. Sample data set and One-Sample Kolmogorov-Smirnov Test of particle mass concentration data.

	Particulate matter particle mass concentrations (µg/m^3)		
	PM 1	PM 2.5	PM 10
N	7520	7520	7520
Normal Mean	3.4533603043E1	1.9541059012E1	1.57282079608E1
Skewness	2.635	2.672	2.183
Std. Error of Skewness	0.028	0.028	0.028
Std. Deviation	2.12527045991E1	1.52710670163E1	1.445027102125E1
Kolmogorov-Smirnov Z	11.201	16.683	17.329
Asymp. Sig. (2-tailed)	.000	.000	.000

Since skewness value is higher than zero and is two times higher than the standard error of skewness, it can be said that the data set is sloping to the right.

In table 4.3. a significance level of 0.00 shows that mass concentration distribution is not normally distributed. In order to see if mass concentration data is distributed log-normally, particle mass concentrations converted to logarithmic concentrations and the one-sample Kolmogorov-Smirnov test is applied. Skewness values are found near 0 which shows distribution maybe log-normal. As a result of the test Asymp. Sig. (2-tailed) the value which is also known as the p-value is higher than 0.05 which means distribution can be accepted as log-normal. Log-normal distribution of PM10, PM2.5, and PM1 mass concentrations are given in figure 4.3.

Table 4.4. Sample data set and One-Sample Kolmogorov-Simirnov Test of logarithmic particle mass concentration data.

	Particulate matter particle mass concentrations (µg/m^3)		
	Log (PM 1)	Log (PM 2.5)	Log (PM 10)
N	7520	7520	7520
Normal Mean	1.4976	1.2007	1.0708
Skewness	-0.268	0.374	-0.157
Std. Error of Skewness	0.028	0.028	0.028
Std. Deviation	0.25385	0.26719	0.31907
Kolmogorov-Smirnov Z	4.838	5.296	4.248
Asymp. Sig. (2-tailed)	.177	.094	.081

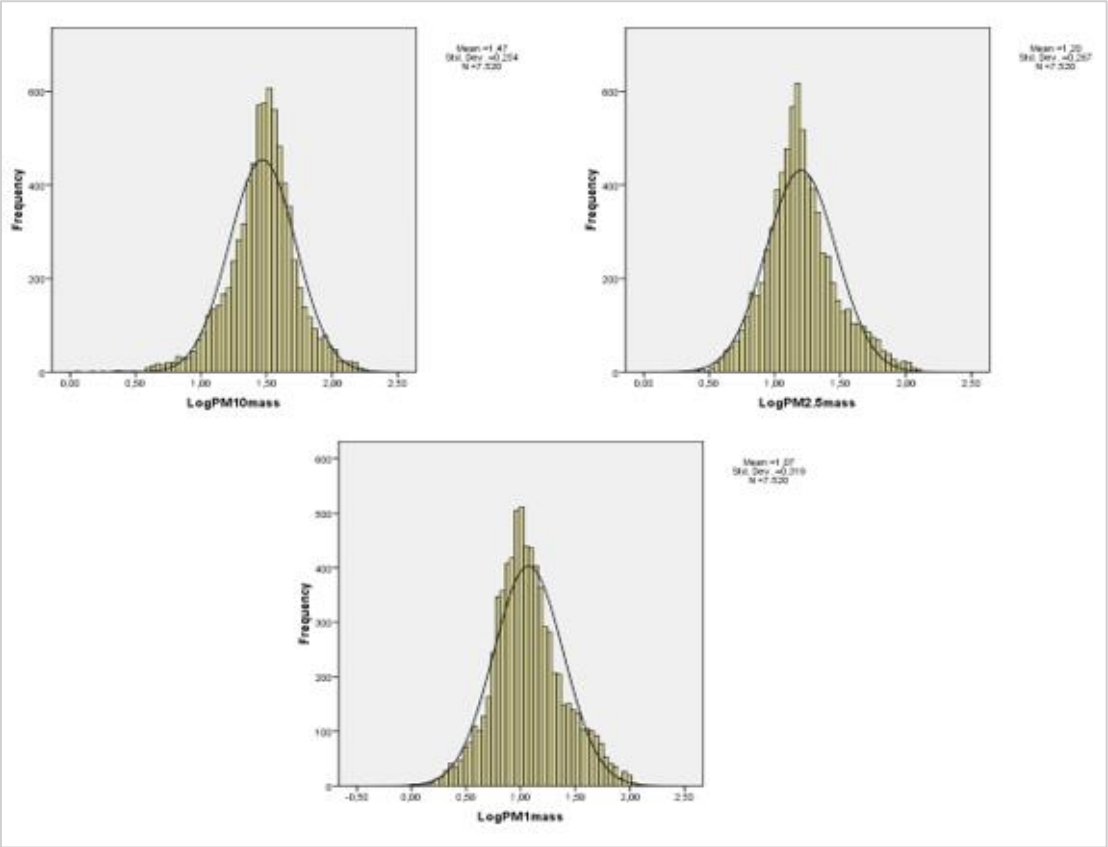

Figure 4.3. Log-normal distribution of particle mass concentration.

4.2. Converting number concentrations into mass concentration

Particle number concentrations are converted to mass concentrations in order to define the distribution of both mass and number concentrations and how they vary with different factors such as temperature, humidity and since regulations are based on mass concentration data number concentration data conversion is necessary for comparison. Multiple regression analysis is used to calculate the mass concentration from number concentration by including the effects of temperature, humidity, and pressure.

GRIMM Ambient Aerosol Monitor with Integrated Sampling Pipe Heater Model 265 device uses a theoretical mass equation to calculate mass concentration based on the particle count concentrations. GRIMM Ambient Aerosol Monitor with Integrated Sampling Pipe Heater Model 265 device gave 4 months of mass concentration data. By using the data of these 4 months and the mass concentration data obtained from stack filter unit, particle count concentration data for each minute,

meteorological data such as pressure, temperature, and humidity equations of PM1, PM2.5, and PM10 are determined with multiple regression.

Pm 10 mass ($\mu g/m^3$) = -68.1891 + 0.0000402561*PM 10 number concentration (count/L) + 0.0798138*pressure (Pa)+ 0.815297*temperature (^0C) - 0.177709*humidity (%rH)

Pm 2.5 mass ($\mu g/m^3$) = 58.1743 + 0.000029567*PM 2.5 number concentration (count/L) - 0.0615758*pressure (Pa) +0.249117*temperature (^0C) - 0.0447557*humidity (%rH)

Pm 1 mass ($\mu g/m^3$) = -22.7107 + 0.0000272974*PM 1 number concentration (count/L)- 0.0243471*humidity (%rH) +0.026955*pressure (Pa)+ 0.0497463*temperature (^0C)

Figure 4.4., 4.5. and 4.6. shows the correlation between the calculated mass with equation versus mass values provided by GRIMM Ambient Aerosol Monitor with Integrated Sampling Pipe Heater Model 265 device. PM 1 an PM 2.5 concentration comparison gave an R^2 value of more than 0.9 which shows that calculated mass concentration is very close to mass concentration calculated by the GRIMM device. PM 10 concentration comparison gave an 0.70 R^2 value even though this result still shows a high correlation between the calculated and GRIMM mass data, it is lower compared with PM 1 and PM 2.5 calculated mass concentration comparisons. This result may be due to other factors that are not considered in this equation such as particulate matter shape estimation, particle volatile and semi-volatile parts, or water absorption capability of particles (Tittarelli et. al., 2008).

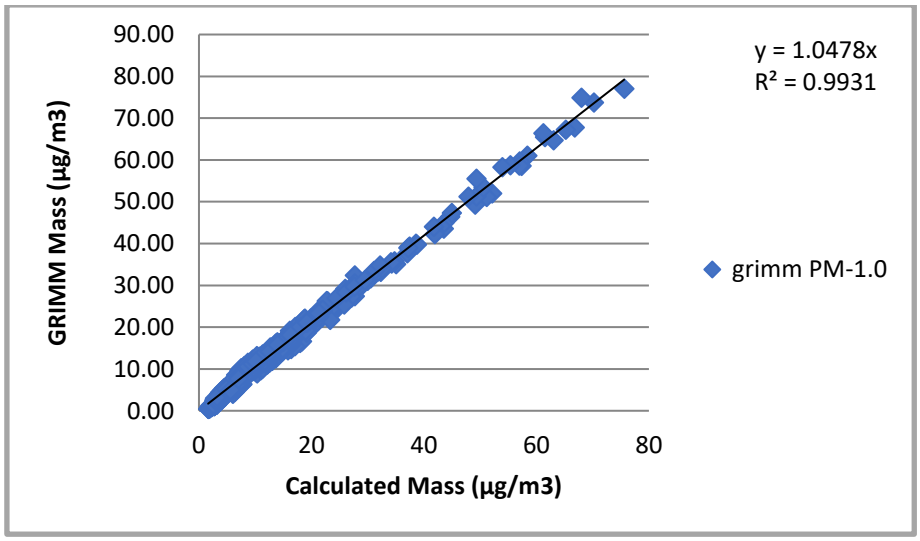

Figure 4.4. PM 1 Calculated mass vs GRIMM mass.

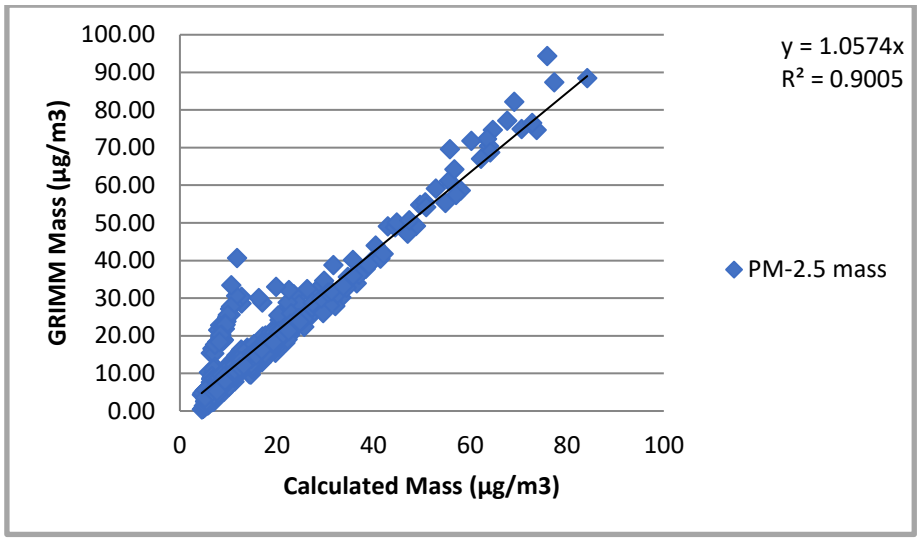

Figure 4.5. PM 2.5 calculated mass vs GRIMM mass.

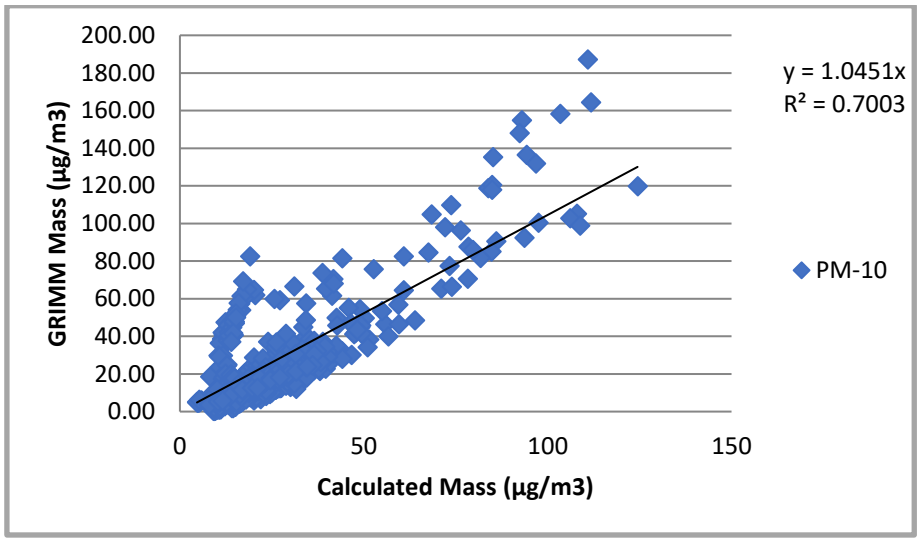

Figure 4.6. PM 10 calculated mass vs GRIMM mass.

4.3. Correlation with meteorology

In order to see the effects of meteorological parameters on particle number and mass concentration, the effect of wind speed, wind direction, temperature, relative humidity, and solar flux is compared with PM 1, PM 2.5 and PM 10 number and mass concentration data. Mass concentration data that are used here are the calculated mass concentration with the previously given equations.

4.3.1. Correlation of number and mass concentrations with wind speed:

The relation of particle mass and number concentrations with wind speed is investigated by simple regression. It is found that wind speed is the most effective factor that changes particulate matter concentration when compared with the effects of meteorological factors such as temperature

and humidity (Çiçek et al., 2004). Strong wind, depending on the mass of particles, moves both particles in the air and the particles suspended. As a result, wind can increase and decrease particle concentration (Kumar et al., 2008). Charron et al., 2003 found that the re-suspension process is for particles larger than 100 nm. On the contrary, an increase in wind speed has no effect on the concentrations of nucleation mode particles (11–30 nm) (Emily et al., 2007).

4.3.1.1. Correlation of particle number concentrations with wind speed. In table 4.5, the distribution of particle count concentration data is given for PM 1, PM 1-2.5, and PM 2.5-10. With this data, figure 4.7 which shows the change of particle count concentration data with wind speed is prepared. For PM 1 with the increasing wind speed decrease in the particle, a decrease in the count concentration is observed which is expected. 1-2 m/s wind speed is found to be the most effective wind speed for the transportation of particles. But for larger particles such as PM 1-2.5 and PM 2.5-10 first, we observe a small decrease in the 1-2 m/s wind speed but with the increasing wind speed concentration distribution is going upward. We observed the peak concentration for PM 1-2.5 at 2-3 m/s wind speed and PM 2.5-10 peak concentration is observed at 4-5m/s wind speed. This movement of particles shows us the effect of resuspension of large particles with high wind speeds.

Another reason for such movement is when wind speed is low, it blows away the pollutants from a certain geographical area but when wind speed increases enough it transports pollutants from other areas that are far away from our sampling location (Wang J. et. al., 2015).

Table 4.5. Change of particle count concentration with wind speed.

Wind speed (m/s)	PM 1 (Count/L)	PM 1-2.5 (Count/L)	PM 2.5-10 (Count/L)
<1	660119.309	2380.583	964.083
1-2	535428.063	2465.065	752.284
2-3	361017.256	2058.183	2340.498
3-4	329166.293	4011.749	1676.426
4-5	280413.594	2932.641	3331.686
5+	235505.081	2408.291	2121.609

In table 4.5, it is seen that with the increasing wind speed particulate matter count decreases as expected. The most decrease in the particle count concentration is observed between the wind speed 2-3 m/s which is nearly half of the previous wind speed sector.

The correlation equation for the change of particle number concentration with wind speed is calculated as in the following based on the figure 4.8 particle count concentration vs. wind speed.

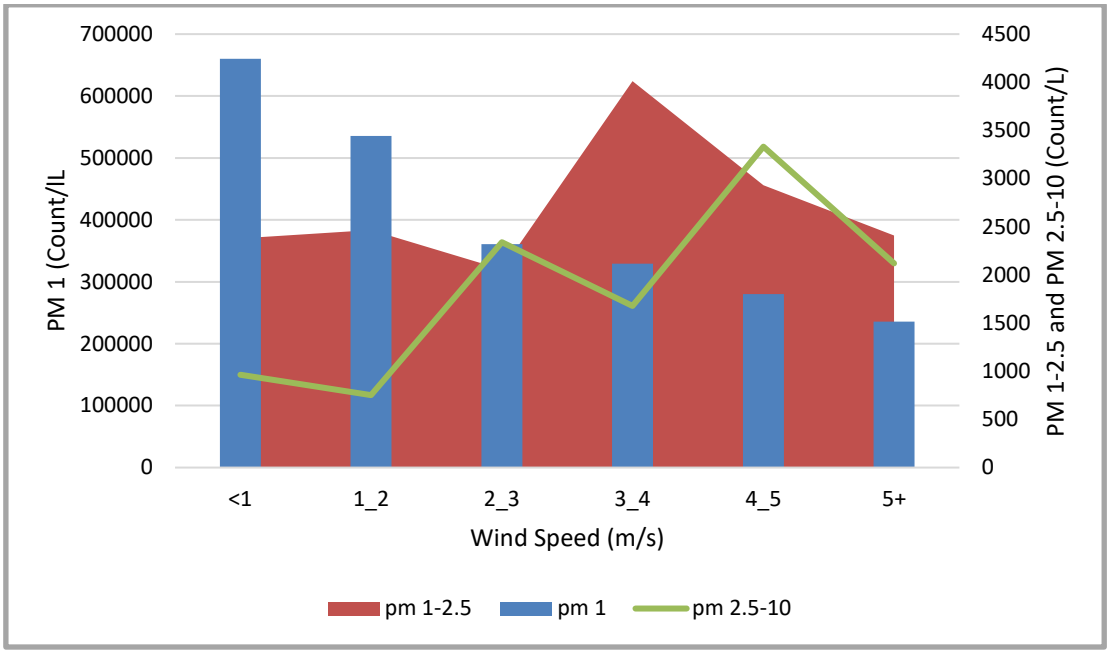

Figure 4.7. Change of particle count concentrations with wind speed.

PM1 (Count/L) = 631830.0 - 81316.2*wind speed(m/s)

PM 2.5 (Count/L) = 634141.0 - 81167.6*wind speed(m/s)

PM 10 (Count/L) = 634925.0 - 80787.6*wind speed(m/s)

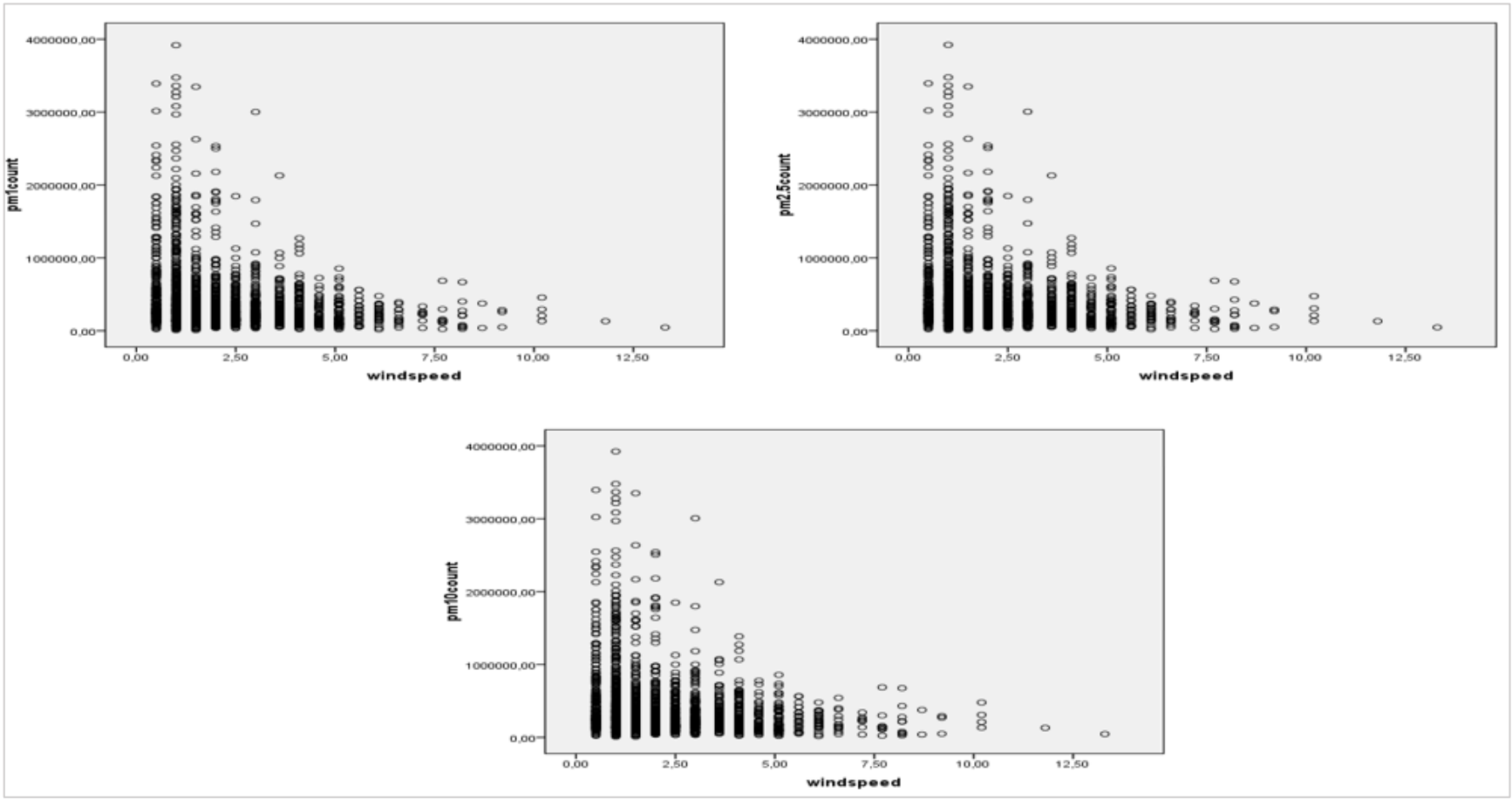

Figure 4.8. PM 1 (Count/L) vs. wind speed (m/s), PM 2.5 (Count/L) vs. wind speed (m/s), PM 10 (Count/L) vs. wind speed (m/s).

4.3.1.2. Correlation of particle mass concentrations with wind speed. In table 4.6, the distribution of particle mass concentration data is given for PM 1, PM 1-2.5, and PM 2.5-10. With this data figure 4.9 which shows the change of particle mass concentration data with wind speed is prepared. For PM 1 with the increasing wind speed decrease in the particle, count concentration is observed until 3-4 m/s wind speed but with the increasing wind speed, PM1 concentration seems to be linear which shows us the small effect of PM1 particle count concentration to mass concentration so we do not observe such a decrease that we have seen in figure 4.7.

For larger particles such as PM 2.5 and PM, 10 similar curves for particle count and particle mass concentration is observed. With the increasing wind speed, first concentration decreases but when speed increases due to resuspension high concentrations are observed. For 3-4 m/s wind speed highest PM1-2.5 count concentration and for wind speed 4-5m/s highest PM 2.5-10 count concentration is observed. For particle mass concentration, the effect of particle resuspension and transportation of particles from other locations is observed (Wang J. et. al., 2015).

Table 4.6. Change of particle mass concentration with wind speed.

Wind speed (m/s)	PM 2.5-10 mass (µg/m^3)	PM 1-2.5 mass (µg/m^3)	PM 1 mass (µg/m^3)
<1	14.865	3.680	17.345
1-2	15.190	3.812	16.880
2-3	14.043	3.638	14.939
3-4	14.453	4.031	14.276
4-5	16.007	3.862	14.122
5<	14.344	3.830	14.116

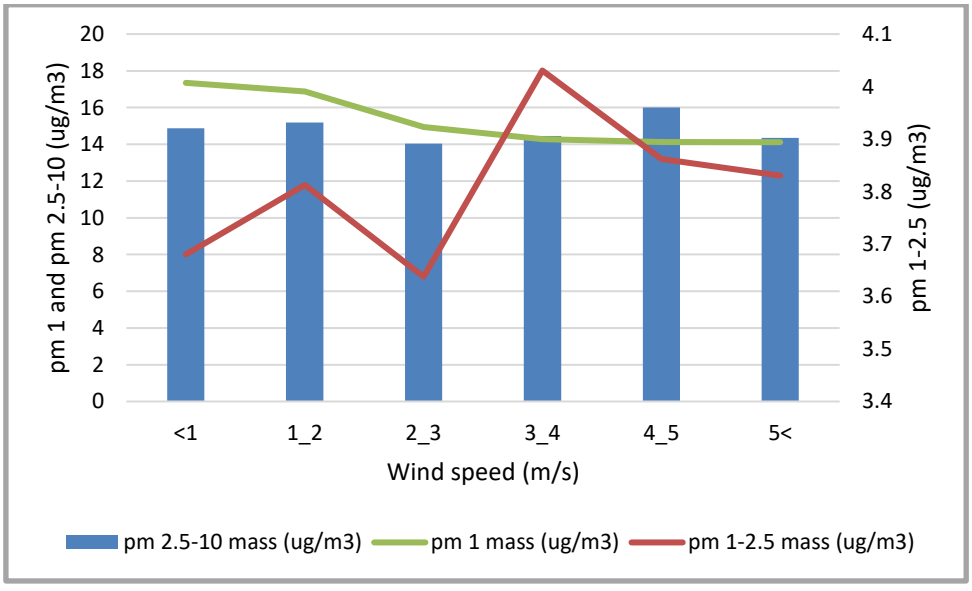

Figure 4.9. Change of particle mass concentrations with wind speed.

The correlation equation for the change of particle mass concentration with wind speed is calculated as in the following based on the figure 4.10 particle count concentration vs. wind speed.

PM 1 mass ($\mu g/m^3$) = 17.2686 - 0.778076*wind speed (m/s)

PM 2.5 mass ($\mu g/m^3$) = 20.7077 - 0.577099*wind speed (m/s)

PM 10 mass ($\mu g/m^3$) = 34.3808 + 0.0988153*wind speed (m/s)

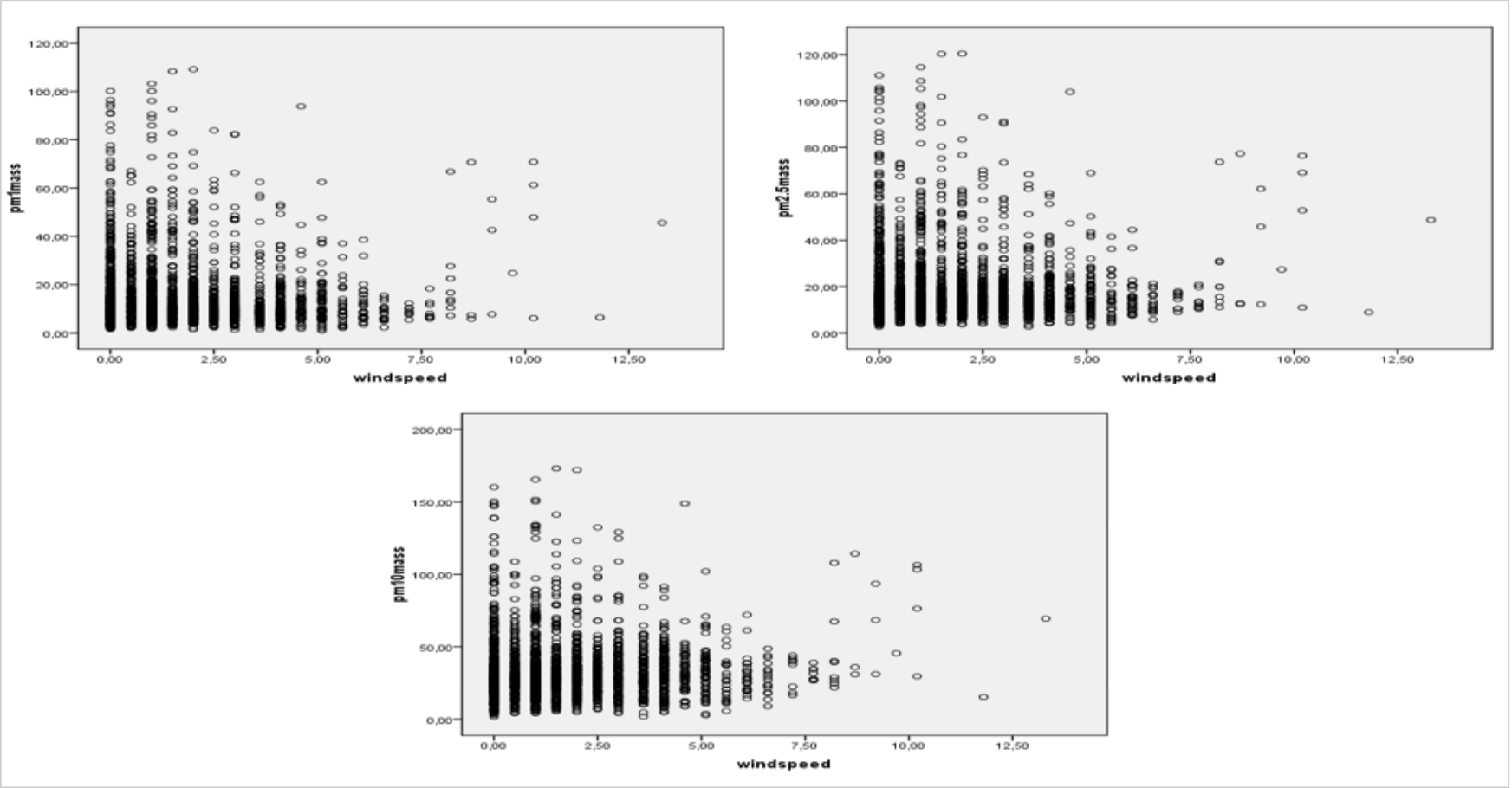

Figure 4.10. PM 1 (µg/m^3) vs. wind speed (m/s), PM 2.5 (µg/m^3) vs. wind speed (m/s), PM 10 (µg/m^3) vs. wind speed (m/s).

4.3.2. Correlation of particle number and mass concentrations with wind direction

Mass and count concentrations of particulate matter are affected by the wind direction since particulate matter may not be distributed homogeneously around the measurement station. It can be understood that if the wind blows from a highly polluted area, measured particulate matter concentration would be high whereas if the wind blows from a cleaner area measured particulate matter concentration would be smaller.

Wind direction data is collected from Etimesgut station and with this data the effect of wind direction on particulate matter mass and count data is calculated. In order to understand the effect of wind direction on particulate matter number and mass concentration, directions are divided into 16 parts which are N, N-NE, NE, E-NE, E, E-SE, SE, S-SE, S, S-SW, SW, W-SW, W, W-NW, NW, N-NW and average concentrations of pollutants are calculated for each wind sector which is named as pollution rose approach.

4.3.2.1. Correlation of particle number concentrations with wind direction. The wind rose graphs for the Etimesgut sampling station during the sample collection period for particle count data with a size of PM 1 particle count (Count/L), PM 2.5 particle count (Count/L), PM 10 particle count (Count/L) is given in Figure 4.11. Prevailing wind direction order changes with some months. But generally, it is seen that prevailing wind direction for PM 1, PM 2.5 and PM 10 particle count concentration is N-NE (north-northeast) and S-SE (south- southeast). The other prevailing wind concentrations can be given as S-SE (south-southeast) and W-NW (west-northwest).

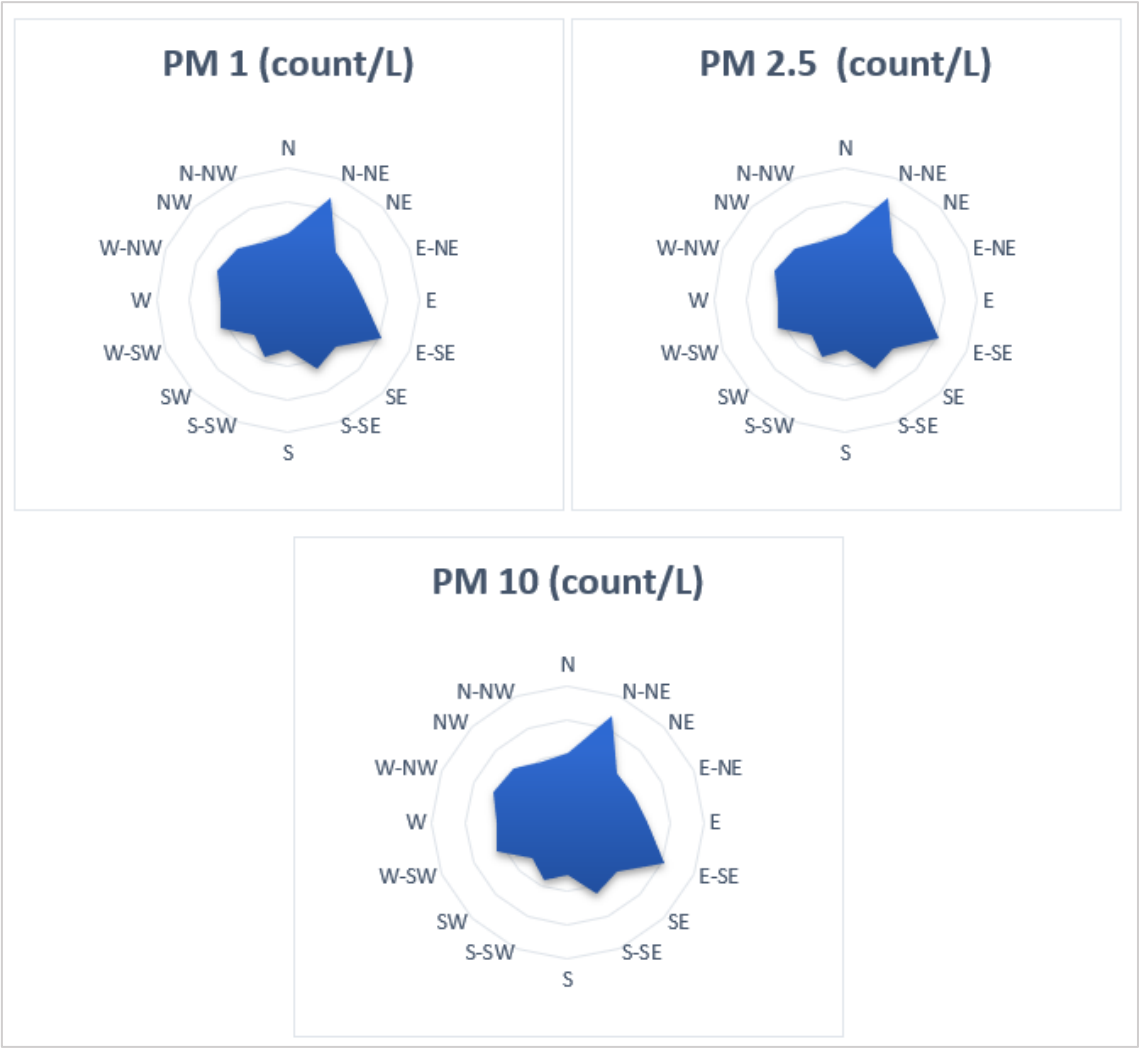

Figure 4.11. Wind direction vs. Particle count PM 1, PM 2.5 and PM 10.

4.3.2.2. Correlation of particle mass concentrations with wind direction. The wind rose graphs for the Etimesgut sampling station during the sample collection period for particle count data with a size of PM 1 particle mass (µg/m3), PM 2.5 particle mass (µg/m3), PM 10 particle mass (µg/m3) is given in Figure 4.12. The ordering of the prevailing wind directions changes with different particle sizes. It is seen that PM 10 particle count concentration does not change much with different wind directions. But generally, N-NE (north-northeast) and S-SE (south- southeast) are seen as the prevailing wind direction for PM 10 particle mass concentration. For the PM 2.5 particle mass concentration data, it is seen that wind direction is more effective than PM 10 mass concentration. For 2.5 particle mass concentration data, generally prevailing wind direction is N-NE (north-northeast) and S-SE (south- southeast). The other prevailing wind directions are W-NW (west-northwest) and W-SW (west-southwest), respectively. For PM 1 particle mass concentration same wind directions of PM 2.5 mass concentration data are observed.

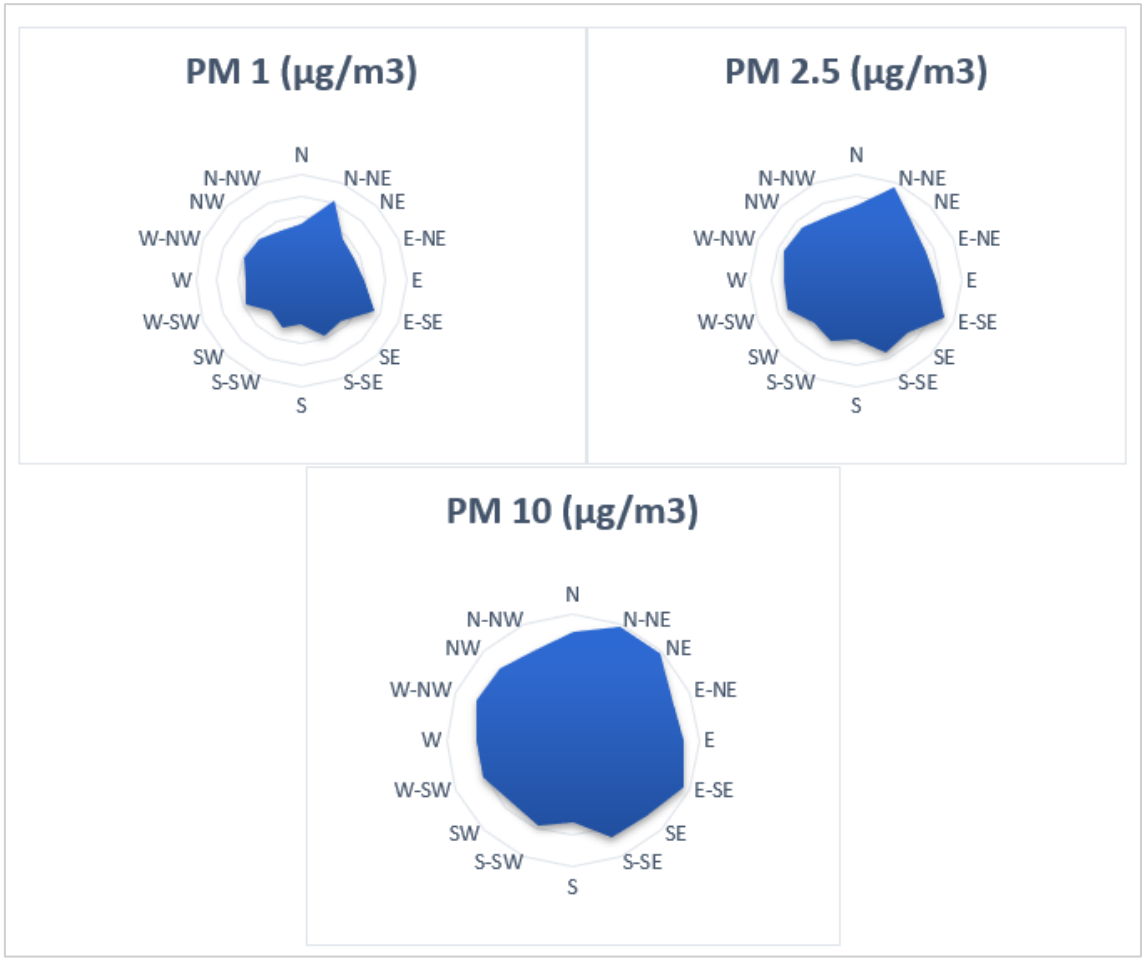

Figure 4.12. Wind direction vs. PM 10, PM 2.5, PM 1 Particle mass concentration

4.3.3. Correlation of particle number and mass concentrations with temperature.

The effect of temperature on particulate matter concentrations is not discussed much in the literature. In research done to find the effect of particulate matter on mortality by Roberts, 2004 it is found that particulate matter is most effective on hot days since hot weather can decrease a person`s physiological response to toxic agents and as a result making them susceptible to the effects of PM. In another study, it is found that with the increasing temperature change, the elder mortality rate is significantly increased (Louis et al., 2013).

In the cold weather, with the increase of household combustion and with the increased fuel usage process, the amount of particulate matter in the atmosphere is increased.

4.3.3.1. Correlation of particle number concentrations for with temperature. In order to see the difference of particle count concentration with temperature, temperature is divided into 8 sectors

which are <-10, (-10)-(-5), (-5)-(0), 0-5, 5-10, 10-15, 15-20 and 20<. Particle count concentrations according to these temperatures are given in table 4.7 and figure 4.13. As can be seen from Table 4.7, peak PM 1 concentration is observed at (-10) - (-5) ^{0}C. Peak concentration for particles between PM 1-2.5 and for PM 2.5-10 is observed at (-5) - (0) ^{0}C. Peak concentrations observed in the cold season is mainly due to fossil fuel combustion for heating. Especially for particles between PM 1- 2.5, an increase with the increasing temperature is observed by reaching the 2nd peak at temperatures above 20 ^{0}C. Since high temperature enhances the photochemical reaction between precursors, such an increase is observed (Wang J. et. al., 2015).

Table 4.7. Particle count concentration change with temperature.

Temperature (^{0}C)	PM 1 count (Count/L)	PM 1-2.5 count (Count/L)	PM 2.5-10 count (Count/L)
<-10	479841.054	275.145	34.25
(-10)-(-5)	968374.601	1069.568	3167.312
(-5)-(0)	883781.649	6802.289	10654.927
0-5	648953.681	3318.338	2414.208
5-10	479421.582	1708.340	386.718
10-15	382954.240	2126.075	512.513
15-20	336677.322	2349.364	624.448
20<	295983.534	3039.749	808.329

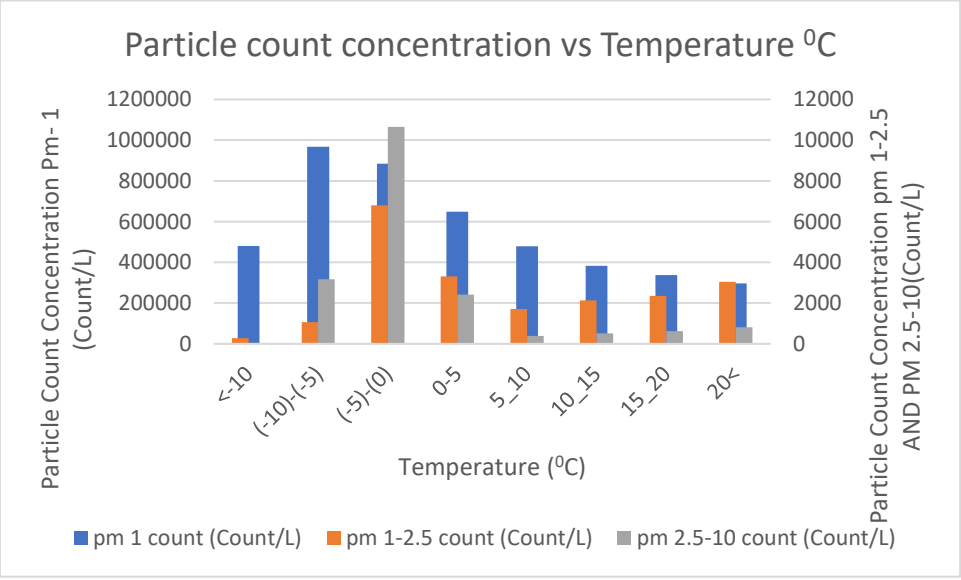

Figure 4.13. Change of particle count concentration with different temperature sectors.

The relation between PM1, PM 2.5, and PM 10 particle count data and temperature is given in figure 4.14. There is a negative relation between particle count and temperature as can be seen. Most of the particle count concentration is observed between temperatures (-10)-(5) 0C. Equation of the

figures is also calculated in order to better understand the effect of temperature on particle count concentration.

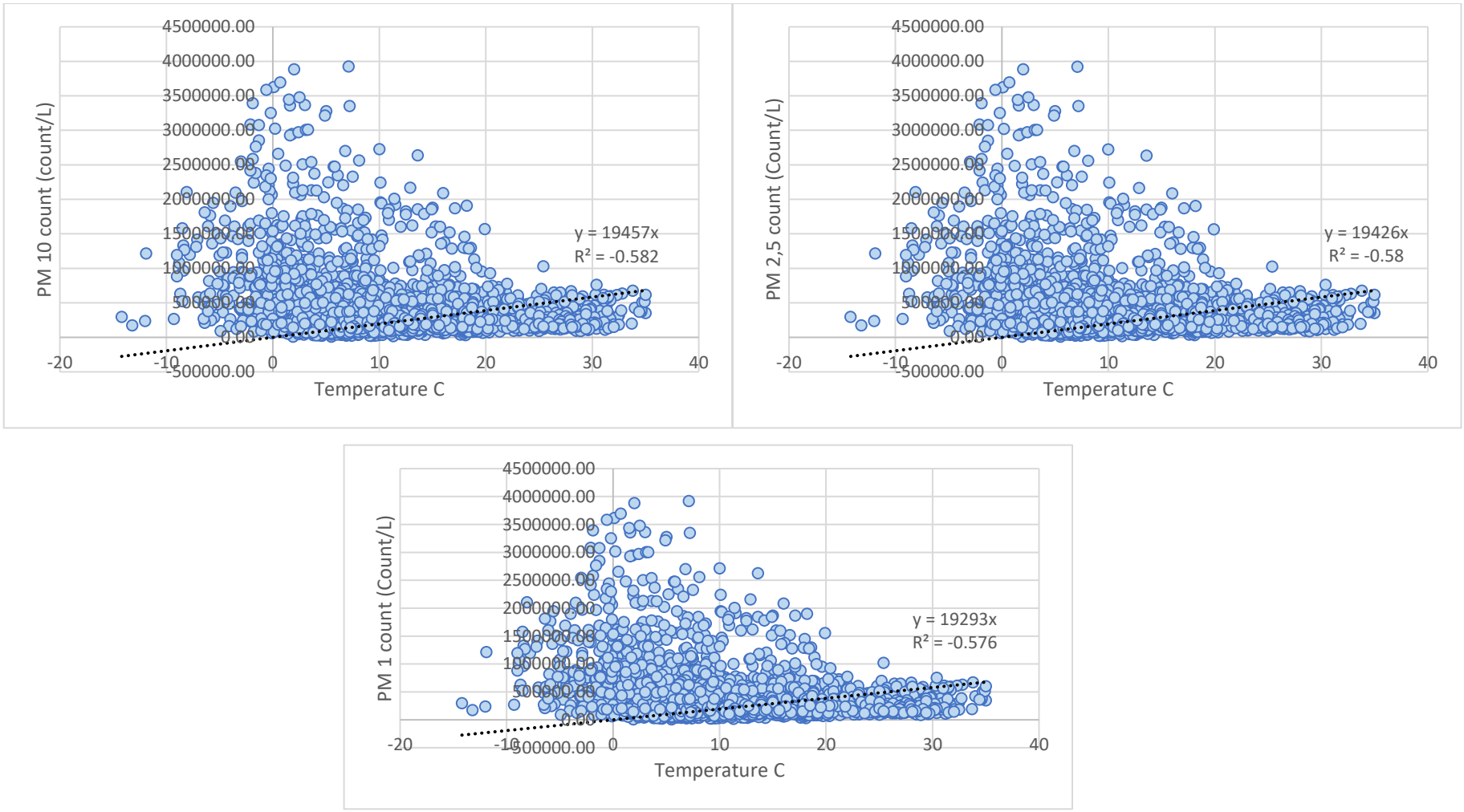

Figure 4.14. PM 1, PM 2.5 and PM 10 count concentration vs. Temperature.

4.3.3.2. Correlation of particle mass concentrations with temperature. In order to see the difference of particle mass concentration with temperature, temperature is divided into 8 sectors which are <-10, (-10)-(-5), (-5)-(0), 0-5, 5-10, 10-15, 15-20 and 20<. Particle mass concentrations according to these temperatures are given in table 4.8 and figure 4.15. As can be seen from Table 4.8, there is a negative relation between the temperature and particulate matter count. The highest mass is also, as expected, observed in the coldest periods. This can be due to the household combustion process. Highest concentration changes are also observed for temperatures -5-0 and 0-5 0C. This shows that these temperatures are the most effective temperatures that affect the mass and count concentrations of particulate matter. In the table, it is also seen that at high temperatures like 15-20 0C, there is an increase in mass concentrations of coarse particles which are seen better in figure 4.16. This result shows the effect of resuspension. On sunny days dust becomes dryer and as a result, resuspension of dust particles is easier since these resuspended particles are large dust particles an increase in the PM 10 mass concentration is observed in hot weather.

Table 4.8. Particle count concentration change with temperature.

Temp	PM 2.5-10 (µg/m^3)	PM 1-2.5 (µg/m^3)	PM 1 (µg/m^3)
<-10	3.552	0.060	14.228
(-10)-(-5)	12.298	1.821	27.843
(-5)-(0)	13.561	2.890	25.557
0-5	11.622	3.115	19.261
5_10	11.991	3.223	14.873
10_15	14.051	3.659	12.628
15_20	16.786	4.273	11.597
20<	22.340	5.597	11.273

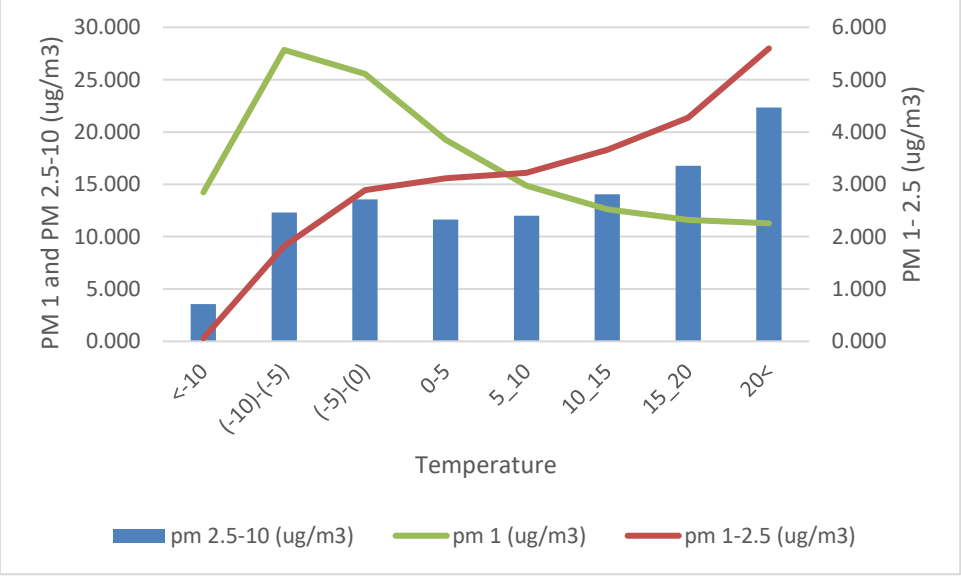

Figure 4.15. Change of particle count concentration with temperature.

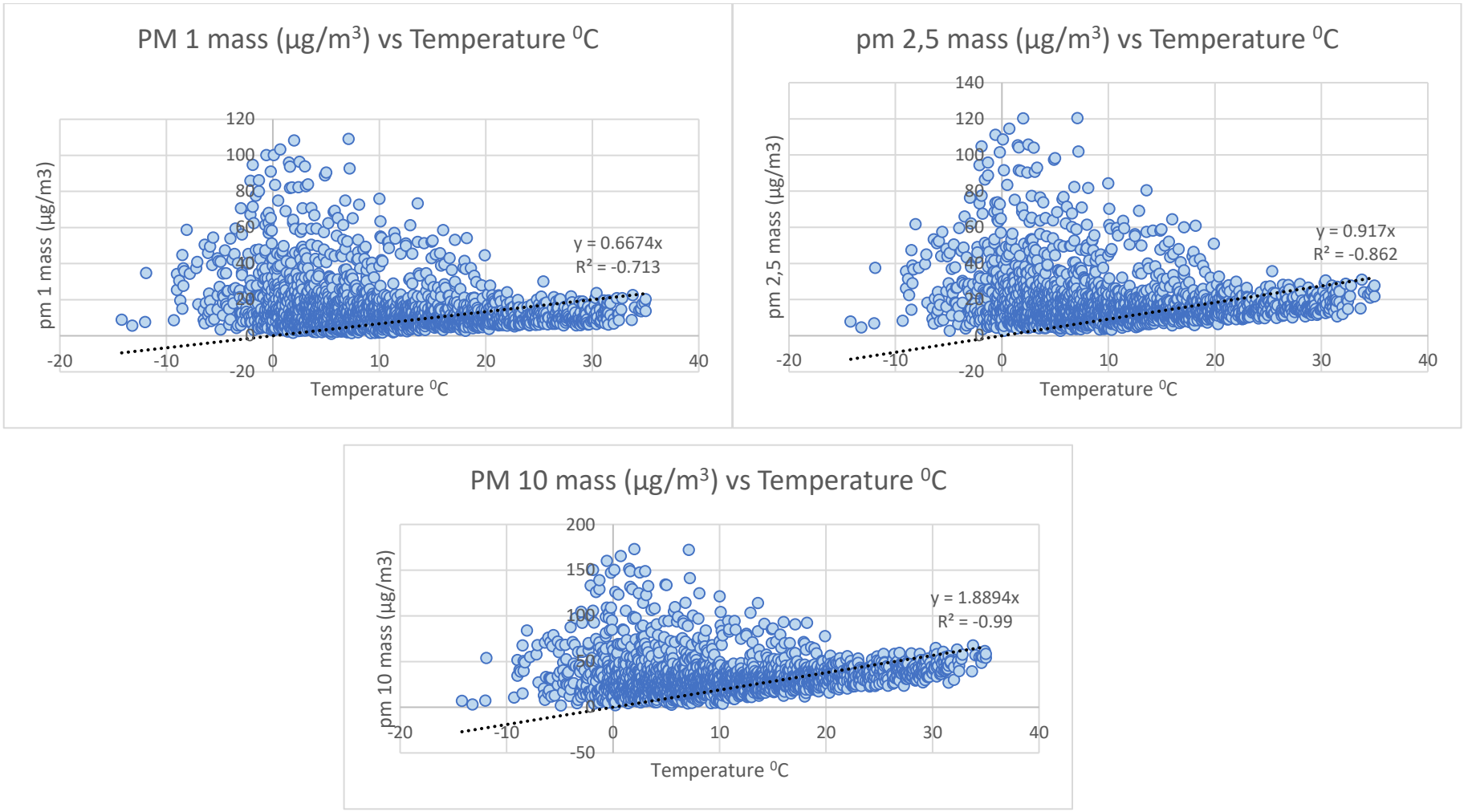

Figure 4.16. PM1, PM 2.5 and PM 10 vs. Temperature.

4.3.4. Correlation of particle mass and number concentrations with relative humidity

Humidity can affect particulate matter sampling. Most of the filters used to determine mass concentration is affected by the relative humidity. Mainly depending on the composition of the PM involved, more mass is found on the loaded filters if the relative humidity is high (Andrew et al., 2006).

Although particulate matter causes respiratory diseases, humidity in the air seems to protect individuals. But if humidity is low, meaning dry air, symptoms may get worse. In research done to determine the effect of particulate matter on chronic bronchitis found that an increase in the relative humidity leads to less hospital admission. This case is found to be correct if particulate matter concentration is less than 60 $\mu g/m^3$. If the concentration gets more than 60 $\mu g/m^3$, the effect of relative humidity on particulate matter concentration is reduced (Leitte et al., 2009).

For humidity comparisons, collected data from the Etimesgut station is analyzed.

The relation between PM 1, PM 1-2.5, and PM 2.5-10 particle count and mass concentration data and humidity is given in figure 4.17 and 4.18, respectively. PM 1 particle count concentration data, increasing particle count concentrations are observed with increased humidity, peak concentration is observed at more than %90 humidity. For larger particles (PM 1-2.5 and PM 2.5-10) first a small increase is observed but with the increasing humidity, a decrease in the particle count concentration data is observed. Highest PM 1-2.5 and PM 2.5-10 is observed at low humidity the reason for that is the hygroscopic growth of particles that increases the concentration. But with the increasing humidity, particles grow too heavy to stay in the air so dry deposition occurs and particles are removed from the atmosphere (Liu, P. et al., 2011). As a result, we observed a decrease in the particle number concentrations with the increasing humidity. Since this dry deposition mechanism does not affect small particles such a decrease is not observed for PM 1 particle count data distribution (Wang J. et. al., 2015).

The relation between PM 1, PM 2.5, and PM 10 particle mass concentration data and humidity are given in the figures 4.19 and 4.20. Increasing particle mass concentrations are observed with increased humidity as seen with the particle count data

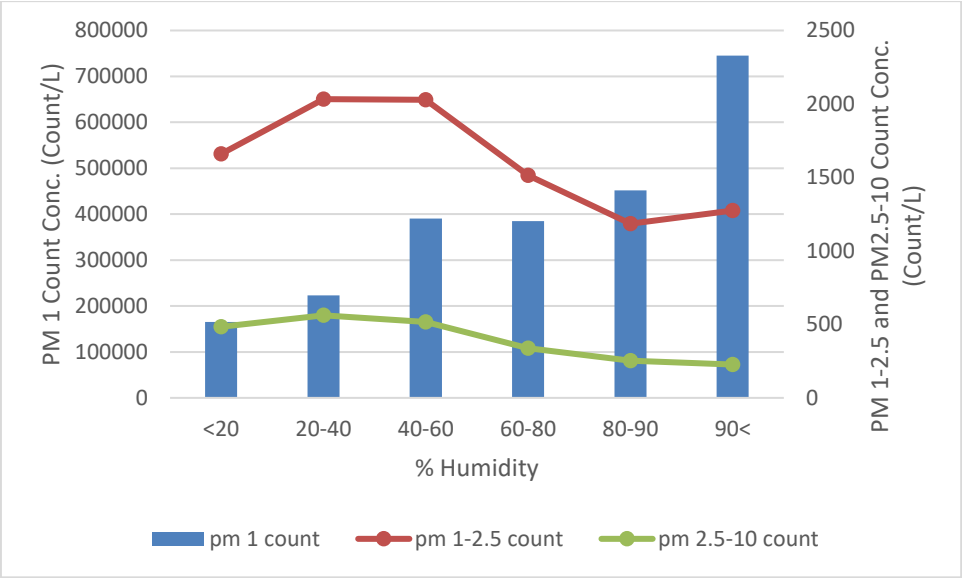

Figure 4.17. Change of particle count concentration with % Humidity.

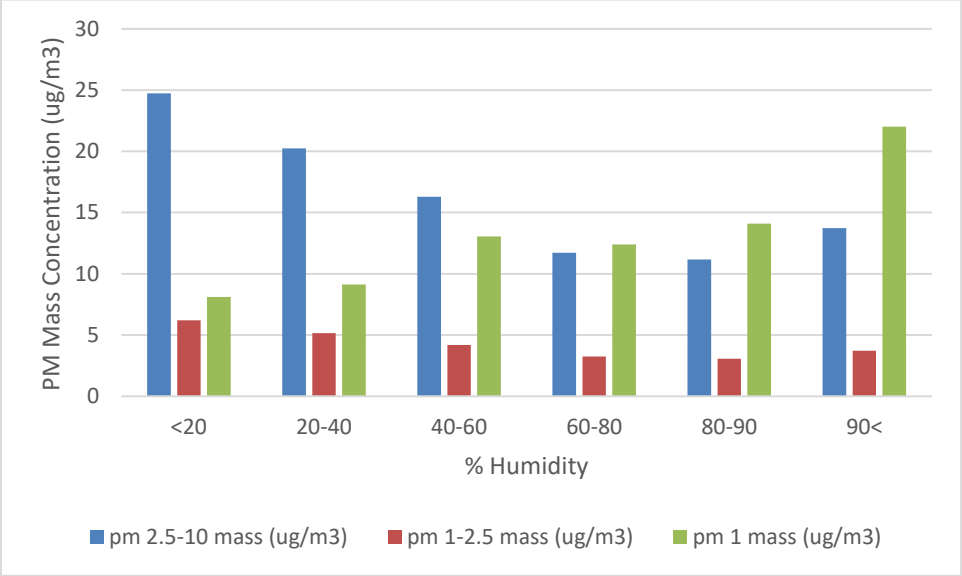

Figure 4.18. Change of particle mass concentration with % Humidity.

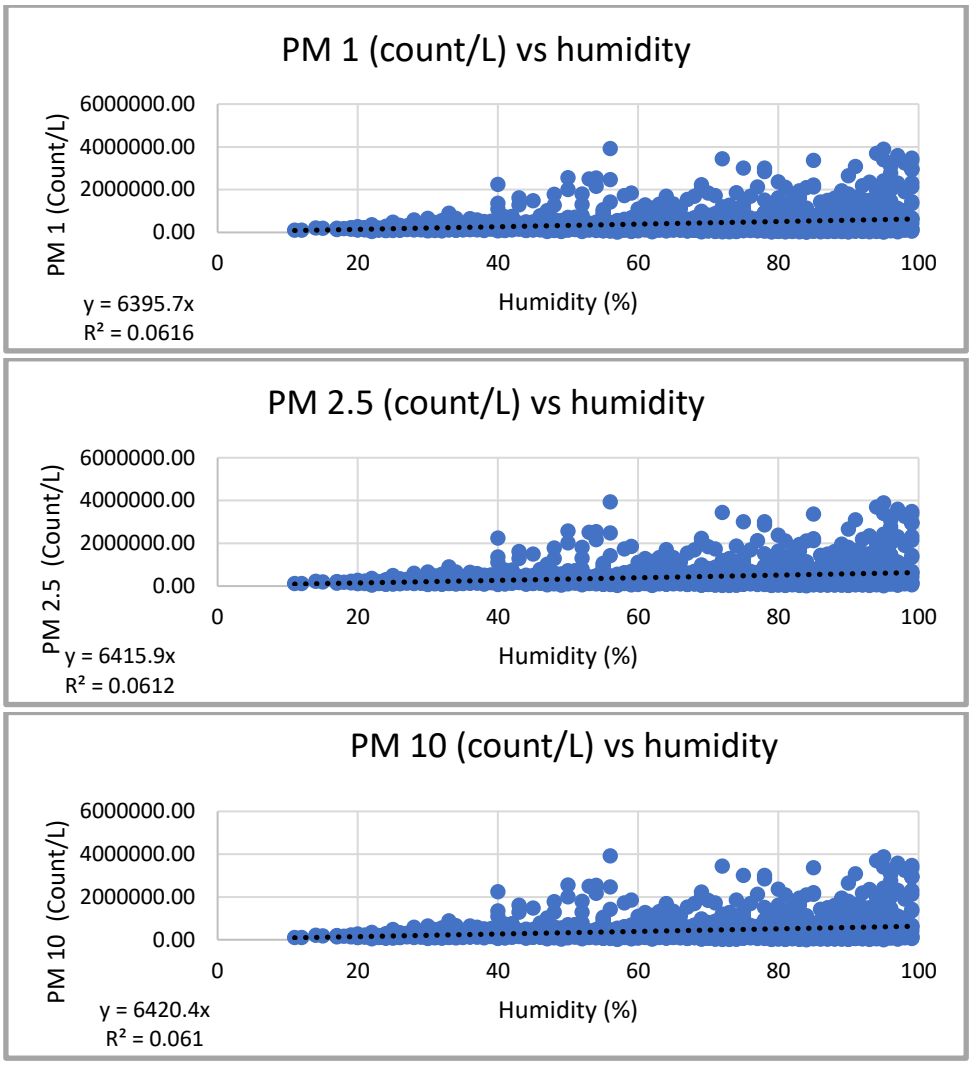

Figure 4.19. PM 1, PM 2.5 and PM 10 number (Count/L) concentrations vs. relative humidity (%).

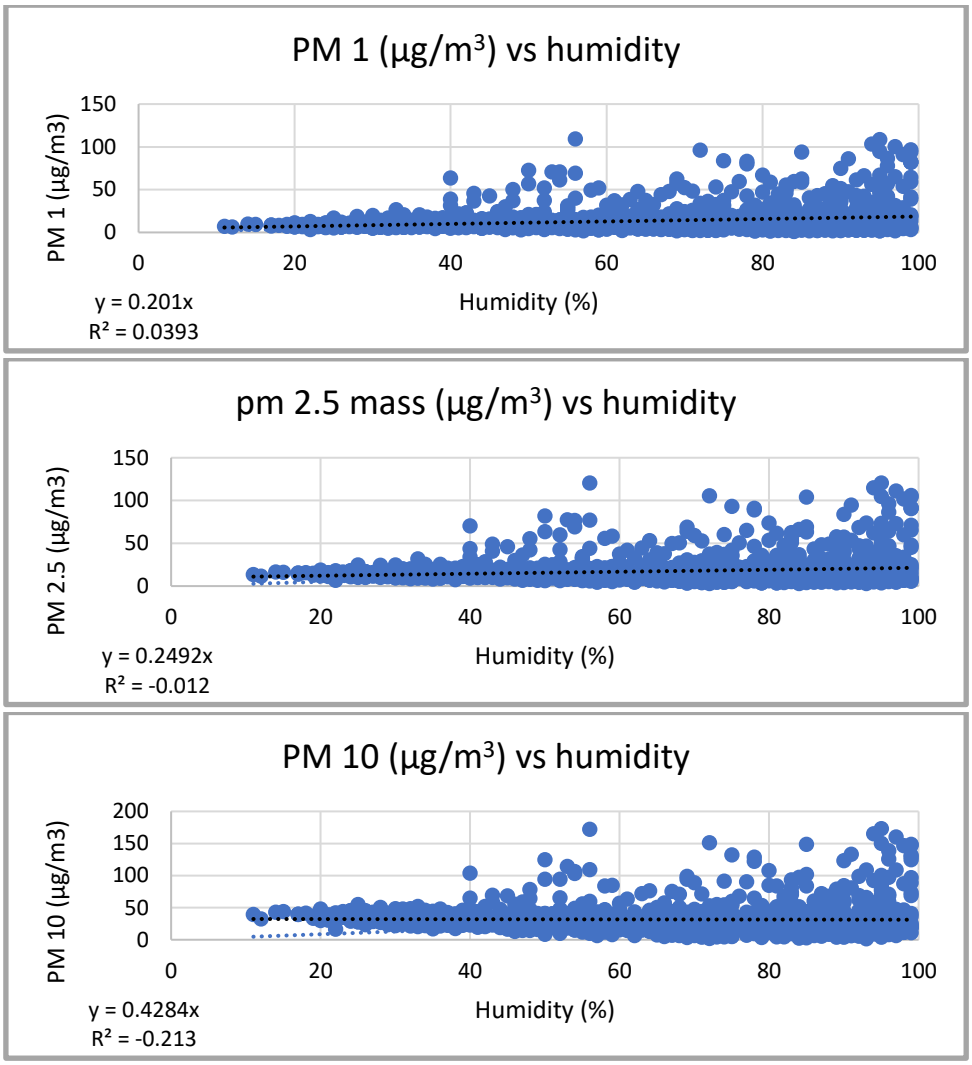

Figure 4.20. PM 1, PM 2.5 and PM 10 mass (Count/L) concentrations vs. relative humidity (%).

4.3.5. Correlation with rain events

As described before, nature helps to remove particulate matter in many ways and one of them is by rain. By absorbing atmospheric gases and trapping particulate matter, raindrops remove pollutants from the atmosphere. In a study, it is found that after the monsoon, there is a considerable drop in the particulate matter concentration also it is found that SO_2 concentration is significantly reduced after rain events (Shukla et al., 2008).

In order to see the effect of rain on mass and number concentration data, daily rainfall data for the sampling time is collected from Etimesgut station. The figure of rainfall days is given in figure 4.21.

Rain days from meteorology:

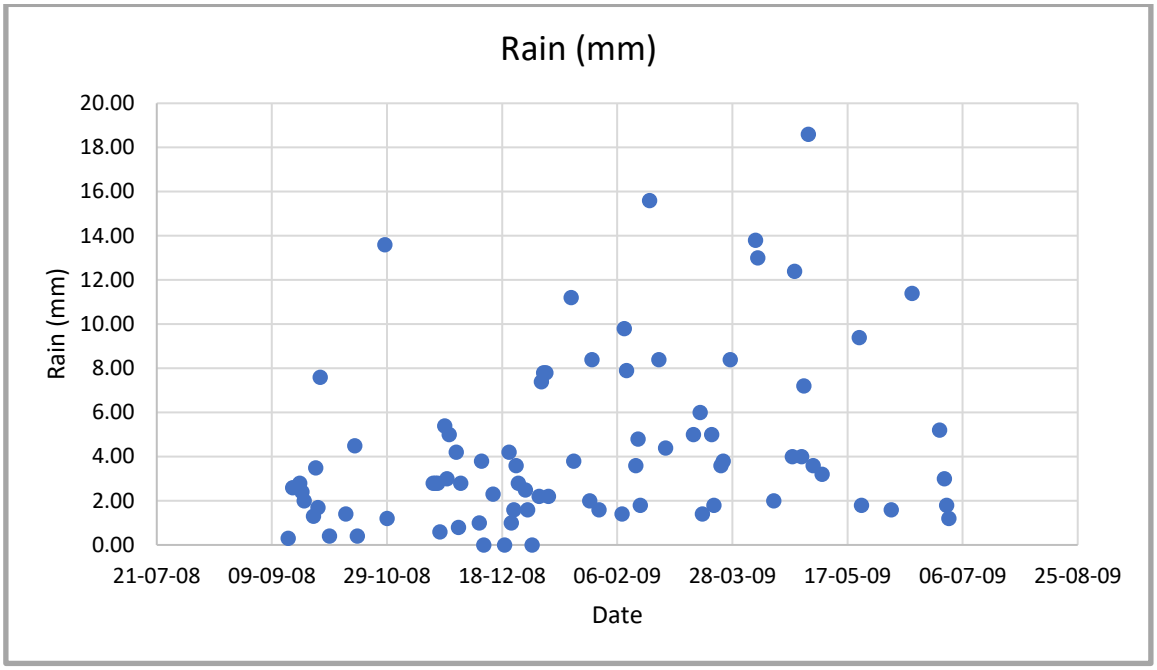

Figure 4.21. Total rain (mm) vs date.

In table 4.9, a comparison of the rainy and non-rainy days is given for both particle mass concentration and particle count concentration. For both particle count and mass data, as expected non-rainy days concentration is much higher than rainy days concentration. This is due to the wet deposition as described before. Particles are removed with rain. For the non-rainy/rainy days ratio of the particle count concentration data, 1.72 ratios are obtained for PM 1, PM 2.5, and PM 10. Since most of the particle count is for PM 1 and less particle count is found for larger sizes, these ratios are nearly equal for different particle sizes. When we look at the non-rainy/rainy days ratio for coarse size particles which are 2.5-10 µm size, we found a ratio of 2.61 which is much higher than other ratios. This result also proves the importance of wet removals effectiveness on large particles.

Table 4.9. Comparison of Rainy days- non-rainy days for particle count and mass concentration.

	Rainy days	Non-rainy days	Non-rainy/rainy days ratio
PM 1 (Count/L)	242873.889	420306.051	1.73
PM 2.5 (Count/L)	244816.552	421297.213	1.72
PM 10 (Count/L)	245101.981	421578.49	1.72
Coarse (Count/L)	171.980767	448.029861	2.61
PM 1 (µg/m^3)	22.2302514	32.529582	1.46
PM 2.5 (µg/m^3)	11.9401771	16.8076782	1.41
PM 10 (µg/m^3)	8.73074719	13.4099345	1.54

In figure 4.22, the size distributions of particles on rainy and non-rainy days are compared. It is seen that for particles with a diameter of less than 1 µm, rainy and non-rainy days average does not change much with rainy and non-rainy days. But for particles with a diameter higher than 2 µm, non-rainy days average concentration is higher than rainy days average concentration for each size bin which also proves the effectiveness of rain on the removal of particles from the atmosphere with wet deposition.

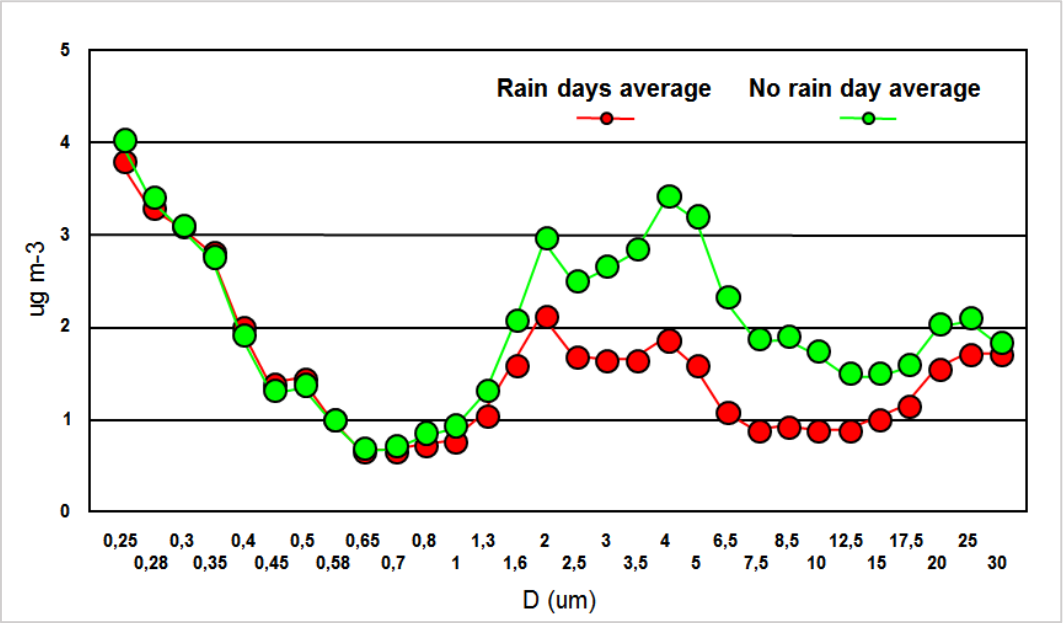

Figure 4.22. Comparison of average rain and non-rain day average size distributions.

Concentrations of the PM 1, PM 2.5, and PM 10 particle count and mass concentration data are sorted according to the days before, during, and after each rain event (figure 4.23 and 4.24). Data is divided as three days before and 10 days after the rain event. In the figures, the main rain event is given as a blue column in the figures. With the main rain event, a significant drop in particle mass and count concentrations is observed which is expected.

Particle count concentration of PM 1-2.5 and PM 2.5- 10 shows an important decrease after the three days' time of a rain event, but this decrease did not observe for PM 1 particle count concentration data. On the other hand, an increase on the 1st day of the rain event for PM 1 particle count data. This difference of movement for different particle sizes with rain days is due to the effective removal of particles with rain and with the following three days of rain event this wet condition continued and a significant decrease in particle count concentration for PM 1-2.5 and PM 2.5- 10 is observed but since as discussed before, rain does not effective on removal of small particles we do not observe such a curve for PM 1. On the 6th and 8th day after the rain event for PM 1-2.5 and PM 2.5- 10 a significant increase of particle count concentration data is observed which is due to loss of effectiveness of rain and particles move to air until removed by dry or wet deposition.

For PM 1 mass concentration data significant decrease is observed in the rain event day but for before and after rain event no specific concentration changes are observed. For PM 1-2.5 and PM 2.5-10 mass concentration data, concentration is increased on the 1st day after the rain event and remained between 15-20 $\mu g/m^3$. For count concentration data of PM 1-2.5, a significant decrease was observed but an increase is observed for mass concentration data. This result showed that although particle number was decreased in the first three days after the rain event, mass concentration is increased. This result is found to be due to the resuspension of large particles which are less in number but high in mass. With the end of the rain event, large particles move to the atmosphere and increase the total mass concentration. A little different figure is obtained for PM 2.5-10 mass concentration distribution with rain events when compared with PM 1-2.5. With the rain event concentration is decreased which is expected on the 1st day after the event, concentration is decreased which shows us the resuspension is low but the 2nd-day concentration is increased, and a decrease is observed in the following days. This result showed us the different characteristics of different size particles and the effectiveness of rain events for each particle size. Different removal mechanisms are effective for different sizes of particles and this difference may be a result of showing such a curve in the figures.

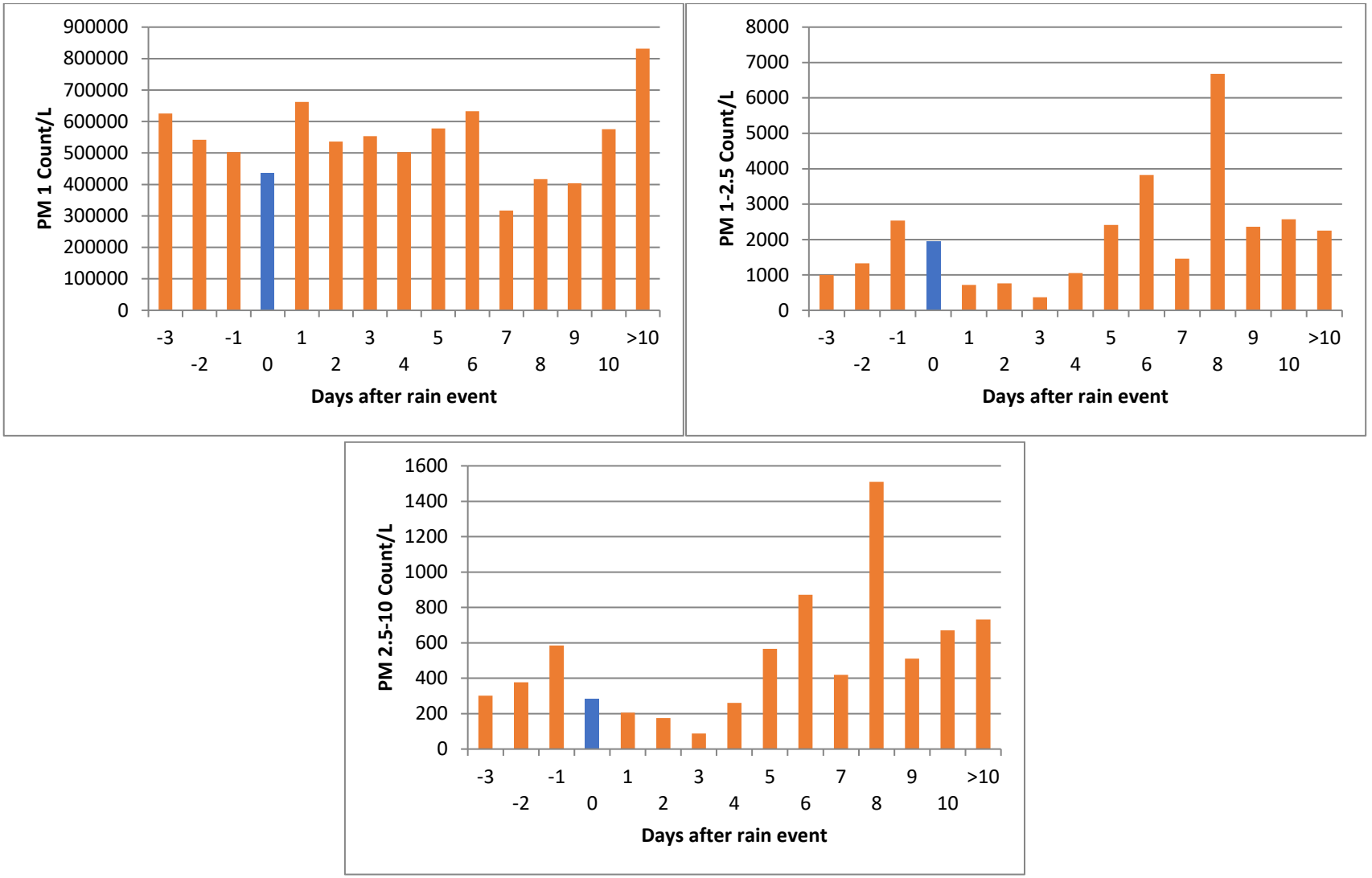

Figure 4.23. PM 1, PM 1-2.5 and PM 2.5-10 particle count data vs. days before and after rain (0 means the day of rain).

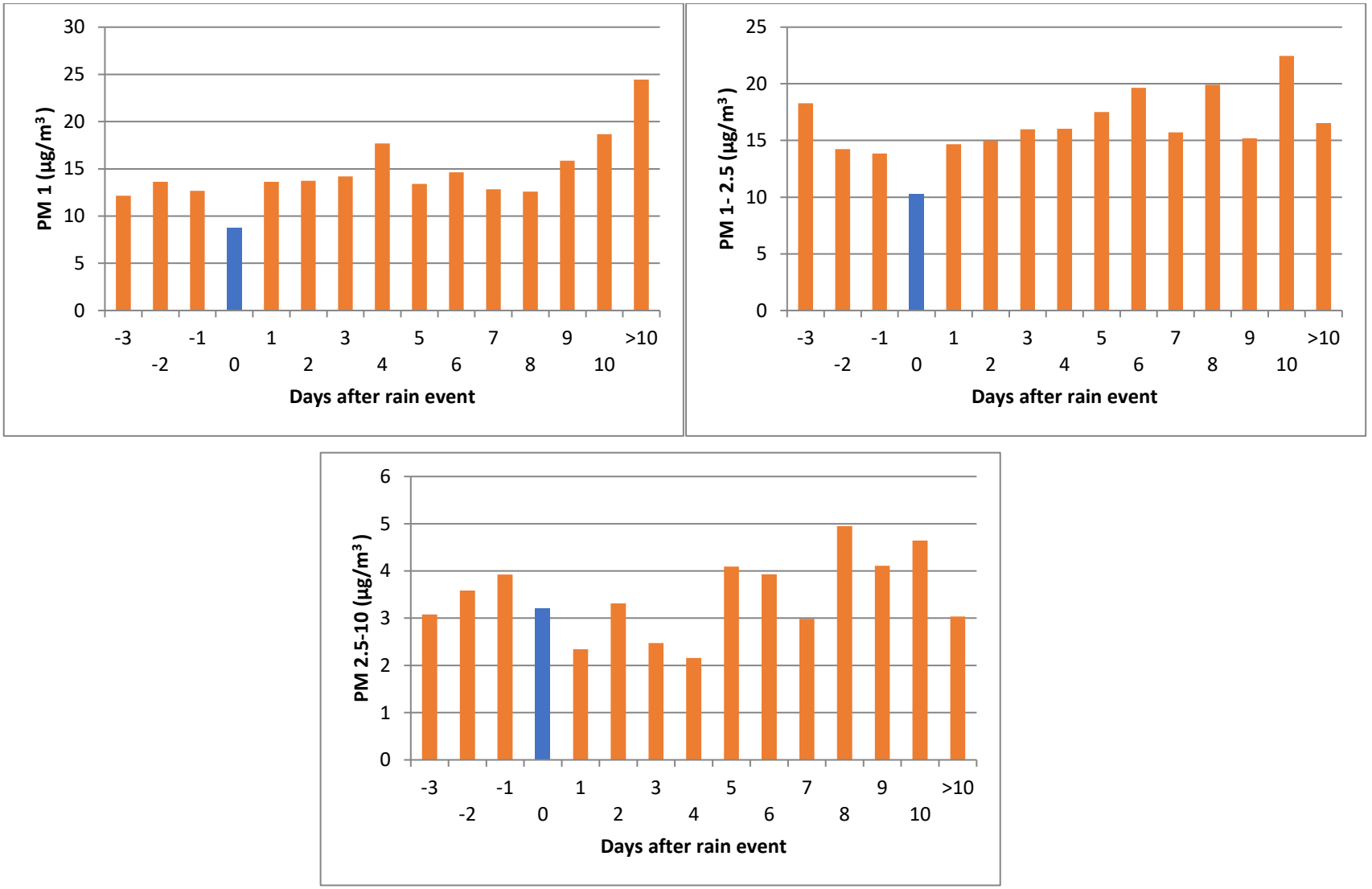

Figure 4.24. PM 1, PM 1-2.5 and PM 2.5-10 particle mass concentration data vs. days before and after rain (0 means the day of rain).

In the following figure (4.25) coarse and fine particle fraction in total suspended particles and their change with rain events are given. It is seen that the fine fraction is high on rainy days and decreases in the latter days when there is no rain event. With the rain event, coarse fraction is decreased but after the rain event fraction of coarse particles in the total mass, concentration is increased. This shows that rain event does not affect all particle sizes in the same way. The effect of meteorology on the removal of particles from the atmosphere is seen better in figure 4.25, coarse-to-fine ratio before the rain event is 0.6-0.7. But when rain event begins, the ratio drops nearly to 0.4. After the rain event ratio continuously increases nearly to 0.8. This profile proves the previous results that rain is much more effective for the removal of particles with a diameter bigger than 2 µm. On the 8th day after a rain event highest coarse to the fine ratio which is nearly 0.8 is obtained that is also parallel to the previous finding for PM 1-2.5 and PM 2.5-10 particle count concentration data distribution.

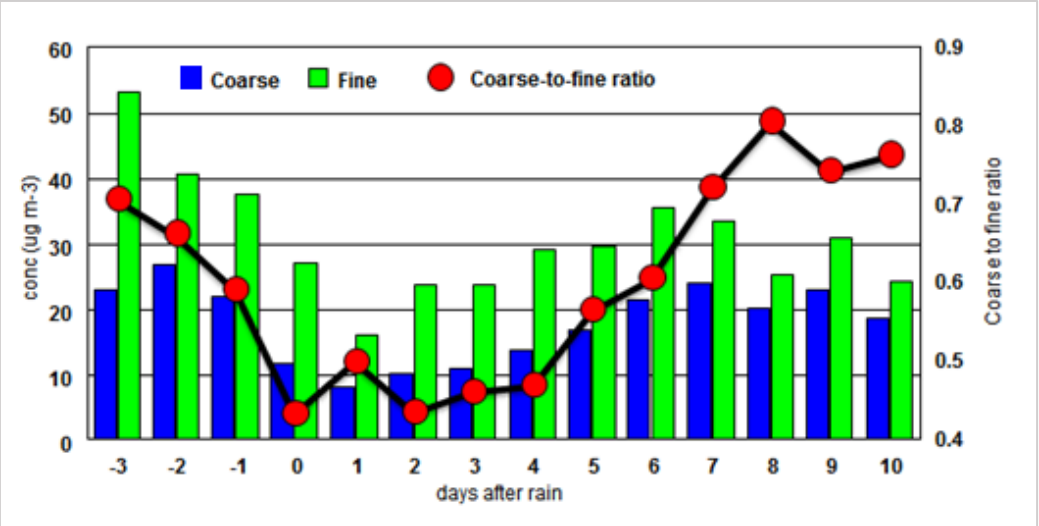

Figure 4.25. Distribution of coarse, fine and coarse to fine ratio after rain days.

4.4. Temporal variation of Particle number and mass concentrations

4.4.1. Diurnal variation of particle count and mass concentration

In order to determine the diurnal change of particulate mass and number concentrations, data of mass and number concentration is divided per hour and average concentrations of each hour are calculated. As a result, figure 4.26 is prepared. In all size fractions, it is seen that concentration is highest between 08.00 – 12.00, and a decrease is observed afterward until 18.00 and after that, there

is an increase again until 22.00. These high concentrations show us the effect of rush hour traffic on particulate matter concentration.

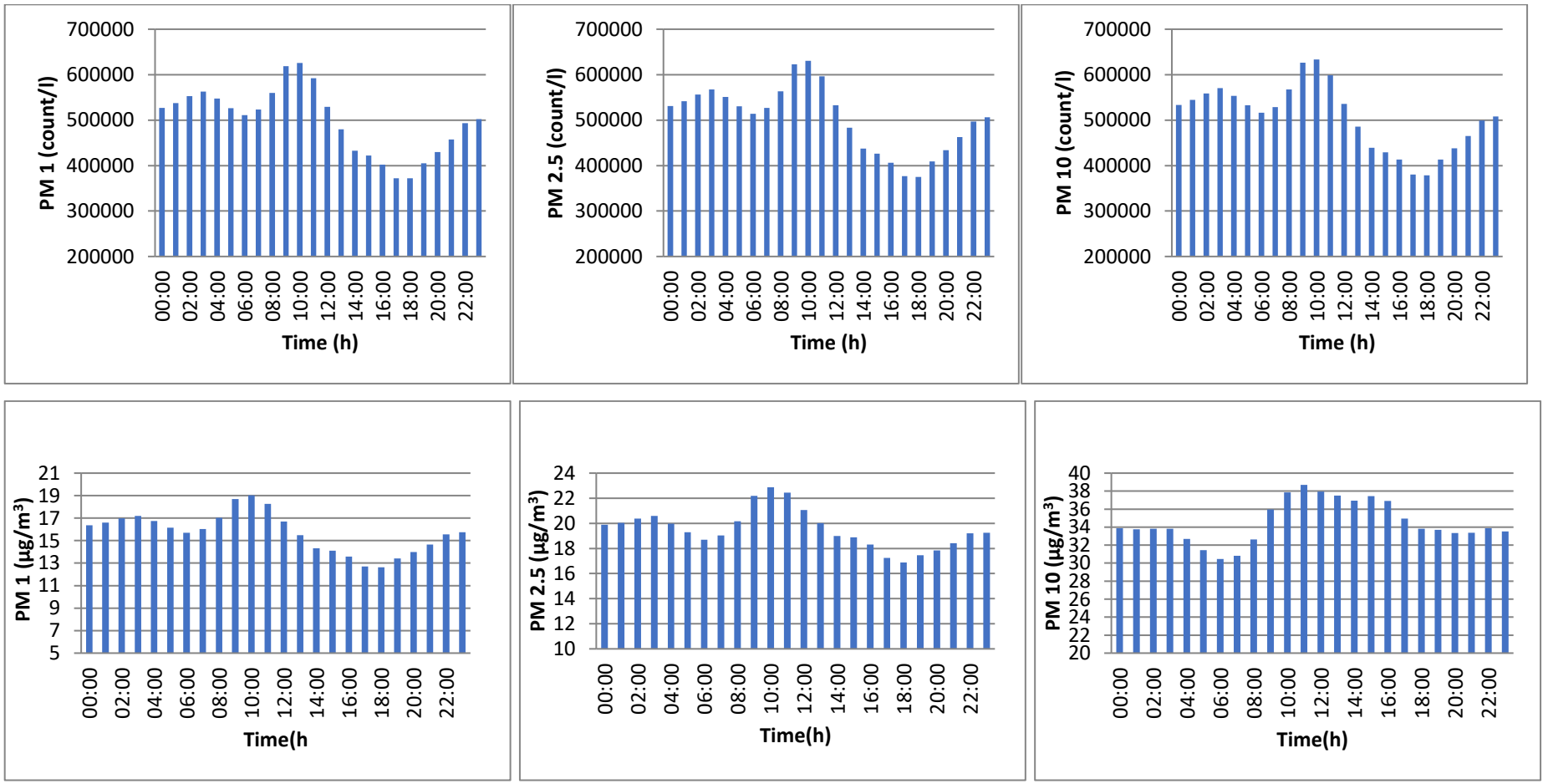

Figure 4.26. Temporal variation of PM 1, PM 2.5 and PM 10 particle count and mass concentration.

In order to understand the diurnal variation of mass size distribution, hourly data of mass concentration is sorted and the average concentration per hour is calculated. In figure 4.27, 2-hour periods are used to minimize disorder. It is seen that at night and early morning times mass concentrations are low. Between 12.00-18.00, there is an increase in the mass concentration but after an 18.00 drop of mass concentration is observed. But this concentration change does not happen in an equal amount for each particle size. In the morning times, fine fraction dominates but after 12.00 fine fraction decreases with an increasing coarse fraction.

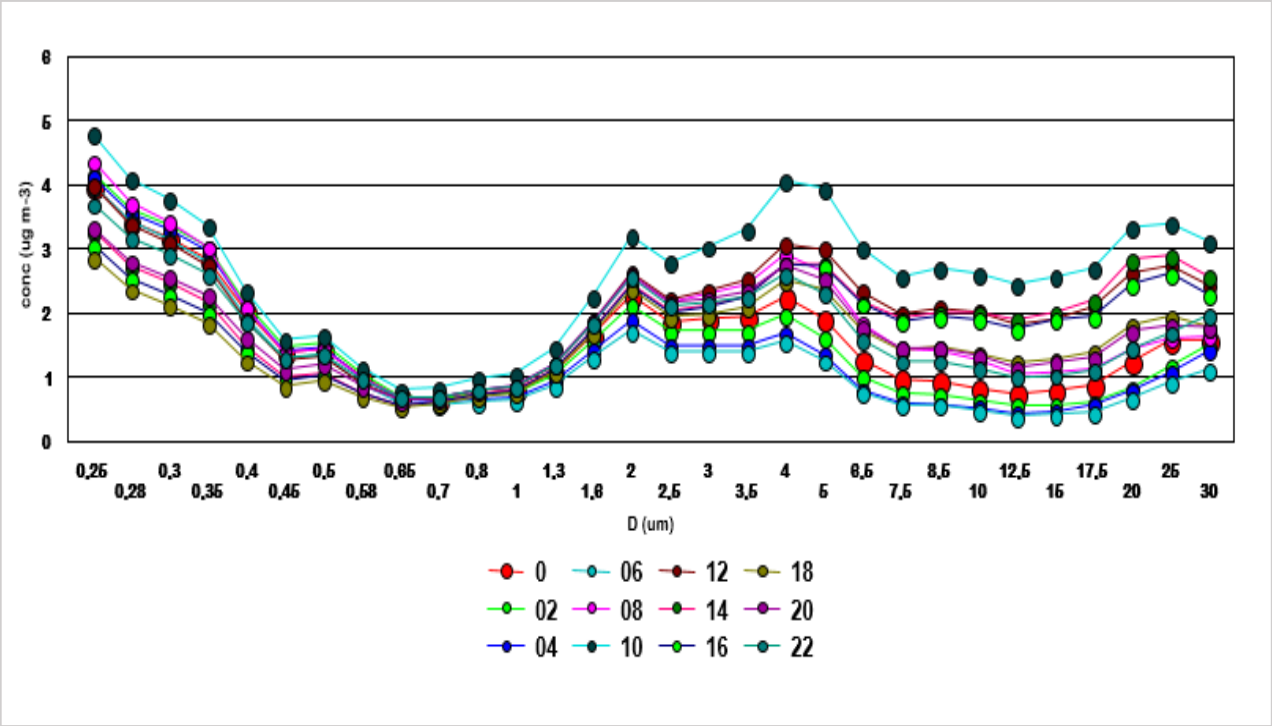

Figure 4.27. Mass size distribution for two-hour periods.

In figure 4.28 coarse, fine, and coarse/fine ratio comparison through a day is given. For coarse and fine fractions nearly, the same profile is observed with the previous figure 4.25 about the PM fractions. As discussed before, particulate matter mass concentration changes with a different rate for different sized particles, and it is also given herewith coarse-to-fine ratio. At night and early in the morning C/F ratio is significantly low and there is a decrease in C/F ratio at night. At night coarse particles settle down but fine particles that are more resistant to settling lead to low C/F ratio. Generally, it is expected for C/F ratio to decrease at rush hours (08:00-11:00) since particles emitted to the atmosphere at these hours assumed to be traffic generated and it is expected for a fine faction to increase and as a result, C/F ratio should decrease. In our case, an increase at the C/F ratio is

observed even rush-hour loses influence after 11:00. This increase and decrease in the C/F ratio show that particulate matter does not only affected by traffic emissions.

There are two kinds of emissions at rush hours. The first one is traffic emissions from car exhausts. It can be seen from figure 4.28, in the rush hours there is an increase in the fine fraction. Second is the resuspension of particles as a result of the movement of the car. The resuspended particles are mostly coarse particles and with the car movement, these particles move to air until another rain event or other removal mechanisms. Since daily activity continues after 11:00, dust continues to resuspend, and as a result decrease in the C/F ratio is not observed. After 19:00, when daily activity decreases, coarse particles settle with a higher rate than fine particles and as a result C/F ratio decreases.

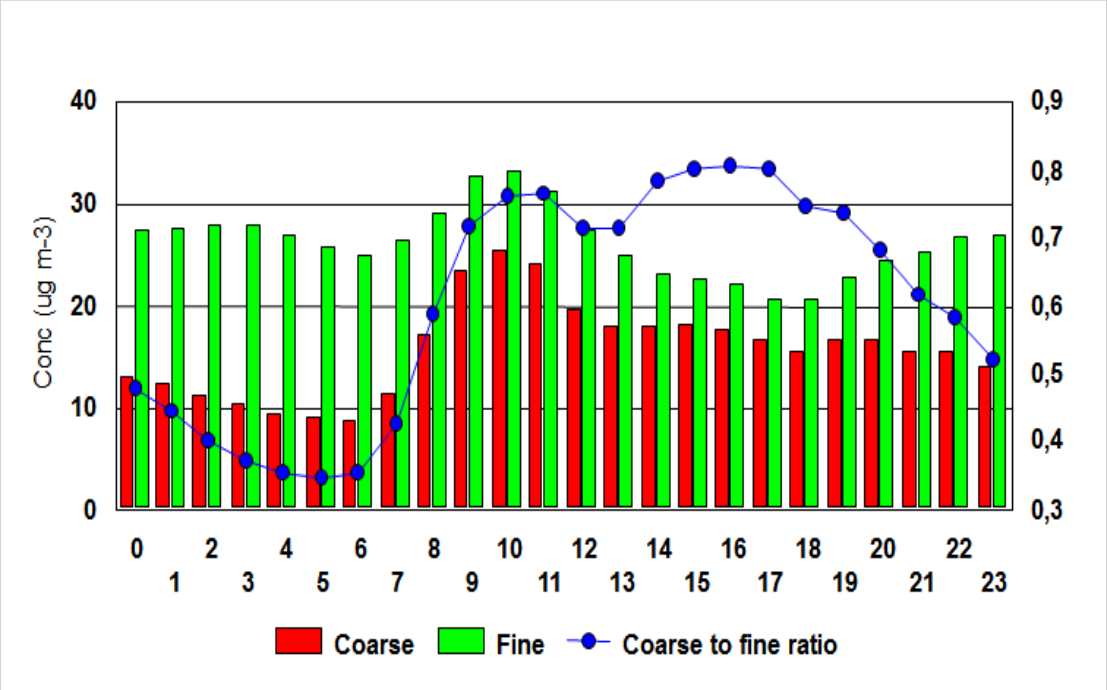

Figure 4.28. Coarse, fine and coarse/fine ratio comparison through a day.

4.4.2. Weekend- weekday variation of particle mass and count concentrations

Weekday and weekend particulate matter concentration comparisons are done in many studies. In a study done in Brisbane, PM 10 mass concentrations are found to be higher during weekdays. (Chan et al., 1997). Many studies showed the same concentration decrease in the weekend for PM 2.5 (Lough et al., 2006). In a study done in Milan, found that PM 10 concentrations show systematic differences between weekdays and weekends. PM 10 concentrations are found to be significantly

lower on Sundays which means the primary source of the particulate matter in the area is traffic (Jones A., 2008). This decrease in the pollutant concentration on weekends is not only caused by the fewer exhaust emissions but also less emission is generated from tire wears and resuspension is low. Also, secondary particles are formed less with decreasing nitrogen oxide emission (NOx). But in some studies, it is found that weekend concentrations of PM 10 do not decrease as expected which means that emissions are not emitted directly from local sources but transported from other sources around the city.

Low particulate matter concentrations are observed at weekends due to holidays of universities and schools, etc. Low car traffic means less emission from tire wears and less resuspension of the dust. Also, since less car traffic means fewer emissions, NOx formation is less observed (Almeida et al., 2005). In the city center, weekend-weekday concentration change is not observed as significant as the urban areas. This shows that particles are not only emitted from local sources but also transported from other areas (Khoder et al., 2008).

Weekend and weekday mass and count concentration data are separated for each season. In table 4.10 and table 4.11, generally, it is seen that there is not much difference between weekend and weekday concentrations. PM 2.5-10 count concentration data is found to be higher in weekdays except for winter season and PM 1-2.5 count concentration data is found to be higher in summer and autumn on weekdays. For particle mass concentration data, in autumn season weekday mass concentration is found to be higher than weekend data.

Table 4.10. Weekday and weekend particle a) count and b) mass concentration for seasons.

a)

	Weekend				
	PM 1 (Count/L)	PM 1-2.5 (Count/L)	PM 2.5 (Count/L)	PM 2.5-10 (Count/L)	PM 10 (Count/L)
Summer	300398.6	1727	302125.6	485.3	302610.9
Autumn	526425.6	2182.5	528608.1	583.9	529192
Winter	760782.3	11154.4	771936.7	11172.7	783109.4
Spring	351375.4	2078.2	353453.6	247.9	353701.5
	Weekday				
	PM 1 (Count/L)	PM 1-2.5 (Count/L)	PM 2.5 (Count/L)	PM 2.5-10 (Count/L)	PM 10 (Count/L)
Summer	255196.9	2201	257397.9	603.6	258001.5
Autumn	524783.8	2519.3	527303.1	694.5	527997.6
Winter	727330.4	8039.8	735370.2	9122.7	744492.9
Spring	368713.6	1549.2	370262.8	351.7	370614.5

b)

	Weekend				
	PM 10 mass (μg/m^3)	PM 2.5-10 (μg/m^3)	PM 2.5 mass (μg/m^3)	PM 1-2.5 (μg/m^3)	PM 1 mass (μg/m^3)
Summer	36.508	20.153	16.355	5.253	11.102
Autumn	35.290	15.215	20.075	3.602	16.472
Winter	38.047	12.365	25.681	3.464	22.216
Spring	28.823	13.625	15.198	3.432	11.765
	Weekday				
	PM 10 mass (μg/m^3)	PM 2.5-10 (μg/m^3)	PM 2.5 mass (μg/m^3)	PM 1-2.5 (μg/m^3)	PM 1 mass (μg/m^3)
Summer	35.360	20.294	15.066	5.107	9.958
Autumn	36.134	15.834	20.300	3.800	16.499
Winter	38.824	13.831	24.993	3.424	21.568
Spring	28.325	12.714	15.610	3.550	12.060

Table 4.11. Weekday/weekend ratio of a) count and b) mass concentration.

a)

Weekday/ weekend	PM1 (Count/L)	PM 1-2.5 (Count/L)	PM 2.5 (Count/L)	PM 2.5-10 (Count/L)	PM 10 (Count/L)
Summer	0.8495	1.2745	0.8520	1.2438	0.8526
Autumn	0.9969	1.1543	0.9975	1.1894	0.9977
Winter	0.9560	0.7208	0.9526	0.8165	0.9507
Spring	1.0493	0.7455	1.0476	1.4187	1.0478

b)

Weekday/weekend	PM 1 (µg/m^3)	PM 1-2.5 (µg/m^3)	PM 2.5 (µg/m^3)	PM 2.5-10 (µg/m^3)	PM 10 (µg/m^3)
Summer	0.8970	0.9723	0.9212	1.0070	0.9686
Autumn	1.0016	1.0550	1.0112	1.0407	1.0239
Winter	0.9708	0.9885	0.9732	1.1185	1.0204
Spring	1.0250	1.0344	1.0272	0.9331	0.9827

4.4.3. Monthly average concentrations of particle mass and number concentrations

All mass and count concentration data are divided into months and averages for each month are calculated. Average monthly concentration changes are given in figure 4.29 and figure 4.30. High concentrations are observed in winter and concentration decreases afterward. PM 1-2.5 mass concentration does not change much for the sampling time. But for other particle sizes, an increase in the mass concentrations is observed from November to January. The lowest mass concentration for PM 10 and PM 2.5-10 is observed in March. For the particle count concentration data, concentrations of PM 1-2.5 and PM 2.5-10 increased sharply in December which may be a result of the long-range transport of particulate matter. Also, the highest concentrations for PM 1 and PM 10 are observed from November to January as seen in the mass concentration data.

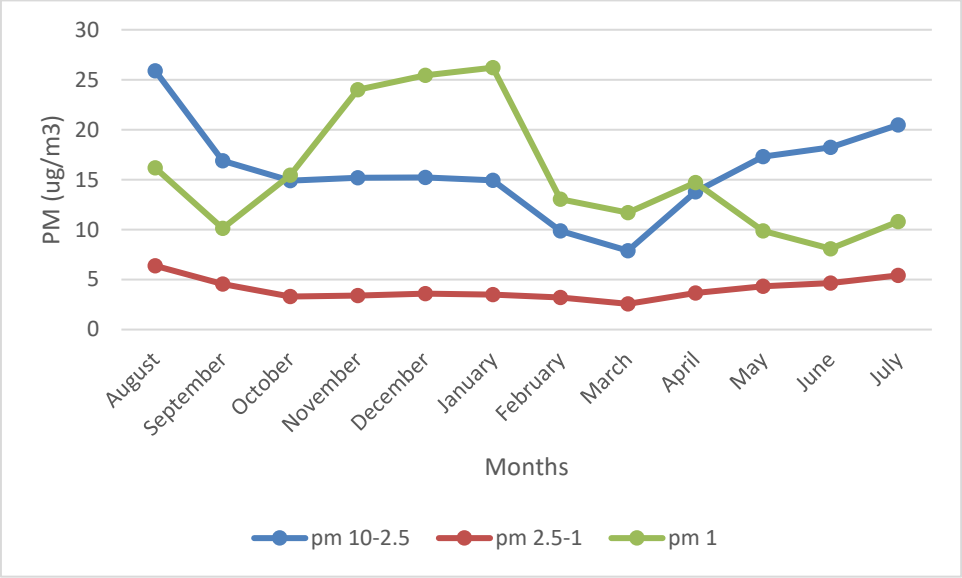

Figure 4.29. Monthly average distribution of particle mass concentration.

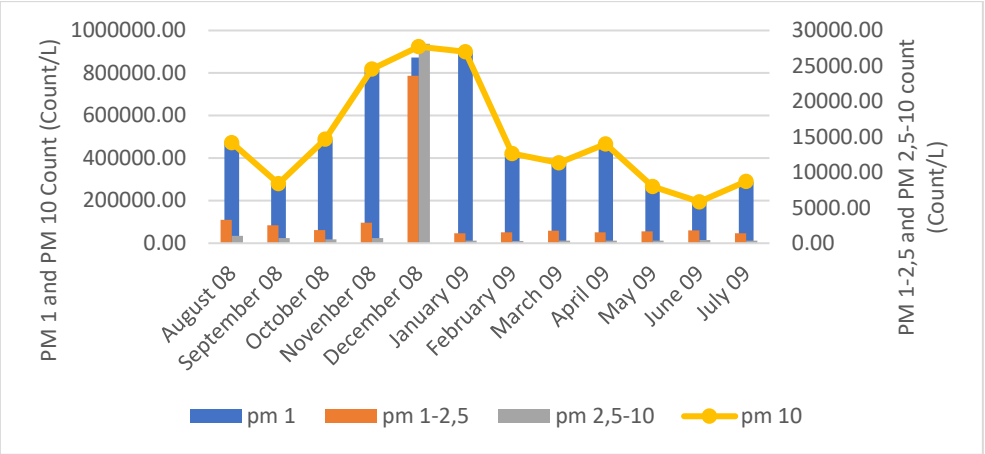

Figure 4.30. Monthly average distribution of particle number concentration.

Winter and summer concentrations distributions with particle size are studied in figure 4.31. It is seen that at winter concentrations of small particles especially submicron particles (D<1 μm) are found to be higher than summer. But large particles (D>5 μm) concentration is lower than summer concentration. For particles between 1-5 μm not much concentration difference between winter and summer. High PM concentration observed in winter is caused by the sources around the city. Anthropogenic emissions as a result of the household heating process are the reason for high PM concentration in winter and since particles emitted to the atmosphere by the combustion process are small-sized, a higher concentration of small particles especially submicron particles (D<1 μm) at winter is observed.

Large particles observed in the Ankara atmosphere in both summer and winter are dust originated particles. These particles can be, as described before, particles resuspended as a result of car movement and dust particles resuspended as a result of wind. The reason for the large dust particles seen higher in summer than winter may be the soil in winter is wet and covered with ice and as a result, dust originated particles are less produced. As a result, it can be said that anthropogenic emissions are the main reason for the high PM concentrations seen in winter, and dominating particle size for the winter season is small particles especially submicron particles (D<1 μm). This result has an important effect. Since, as previously discussed, as the matter particle size becomes smaller, the effect of particulate to the human health increases since particles can penetrate deeper through the alveolar region of the lungs and they can affect other organs (Oberdorster 2001; Branis et al. 2005).

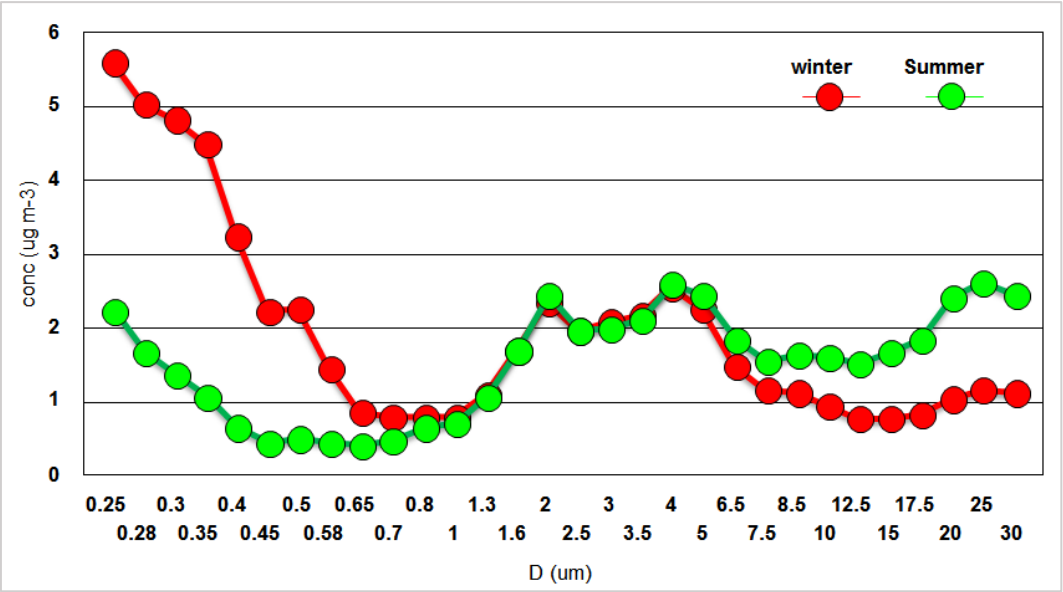

Figure 4.31. Winter and summer concentration distribution with particle size.

4.5. Variation of particle size distribution with dust events

Wind erosion from desert regions is an important soil-derived aerosol source that affects the global aerosol budget in a considerable way (Kaufman et al., 2005). Chemical, physical and mineralogical properties of a mineral determine the effect of a dust particle on climate and atmospheric chemistry (Jeong et al., 2007). Dust from the deserted areas can be transported over long distances with the wind. In a study, it is found that Saharan dust can transport long distances even to the continental area of Northern and Southern America (Formenti et al., 2001) and desert dust from Takla Makan desert is found to eastern China to the pacific coast of America (Makra et al., 2002).

When Saharan desert air masses move northward over the Ankara, Turkey, Saharan intrusion at the study area occur and as a result of the high mineral contentment of Saharan dust, high PM and TSP are observed at the air quality stations.

In a study, it is found that PM 10 concentration before the dust event is found to be low which is as a result of the arrival of Atlantic air masses preceding the northward high particulate flow and during the dust event, there is a sharp increase observed in particulate matter concentrations which is due to the plume-like behavior of the Saharan intrusion. At the end of events, particulate matter concentrations decreased rapidly which is mainly due to rainfall (S. Rodriguez, 2001).

Hourly PM 10 and SO_2 concentration data during the 12 months of particle count concentration sampling time is collected from six stations in Ankara which are Bahcelievler station, Dikmen station, Cebeci station, Kecioren station, Demetevler station and Sihhiye station given in table 4.12.

Table 4.12. Summary statistics of hourly PM10 mass concentrations (in ug/m^3) of each station.

	Bahcelievler PM10	Cebeci PM10	Demetevler PM10	Dikmen PM10	Keçiören PM10	Sıhhiye PM10
Number of valid data	6980	2823	6571	2703	4080	3121
Number of missing data	292	4449	701	4569	3192	4151
mean	54,766	62,204	71,796	43,981	46,469	81,962
std error of mean	0,911	2,049	1,007	0,752	0,855	1,482
median	33,0	39,0	44,0	31,0	31,0	62,0
mode	14,0	8,0	24,0	21,0	0,0	44,0
std deviation	76,123	108,845	81,627	39,088	54,624	82,766
variance	5794,755	11847,2	6662,986	1527,886	2983,731	6850,1
skewness	5,819	23,270	3,237	4,278	5,906	6,915
kurtosis	46,950	873,875	16,381	52,635	67,525	96,13
maximum	980	4368	965	790	961	1679

In order to define dust episodes, particulate matter concentration data from these stations is examined simultaneously. Data with a concentration higher than the average concentration of the station is selected and if the selected data is with a high concentration than average concentration and since the particles reaching to Ankara by long-range transport are expected to be observed in all or most of the stations if it is common to all or most of the 6 stations simultaneously than the data set is selected as candidate dust episode. Also, if the selected period was shorter than 10 continuous hours, it is removed from candidate data. This removal is done in order to reduce local meteorology effects like inversions during the night and before sunrise (Karaca et al., 2009). Afterward, the BSC-DREAM8b (Barcelona supercomputing center) dust model which predicts the atmospheric life cycle of the eroded desert dust is used to see if there is dust transport around Ankara. If the dust transport is observed at the same time with the previously determined candidate episode data than the data set is selected as a dust episode.

In order to find the common dust episodes in the station Distribution of hourly PM 10 concentration data is given in figure 4.32.

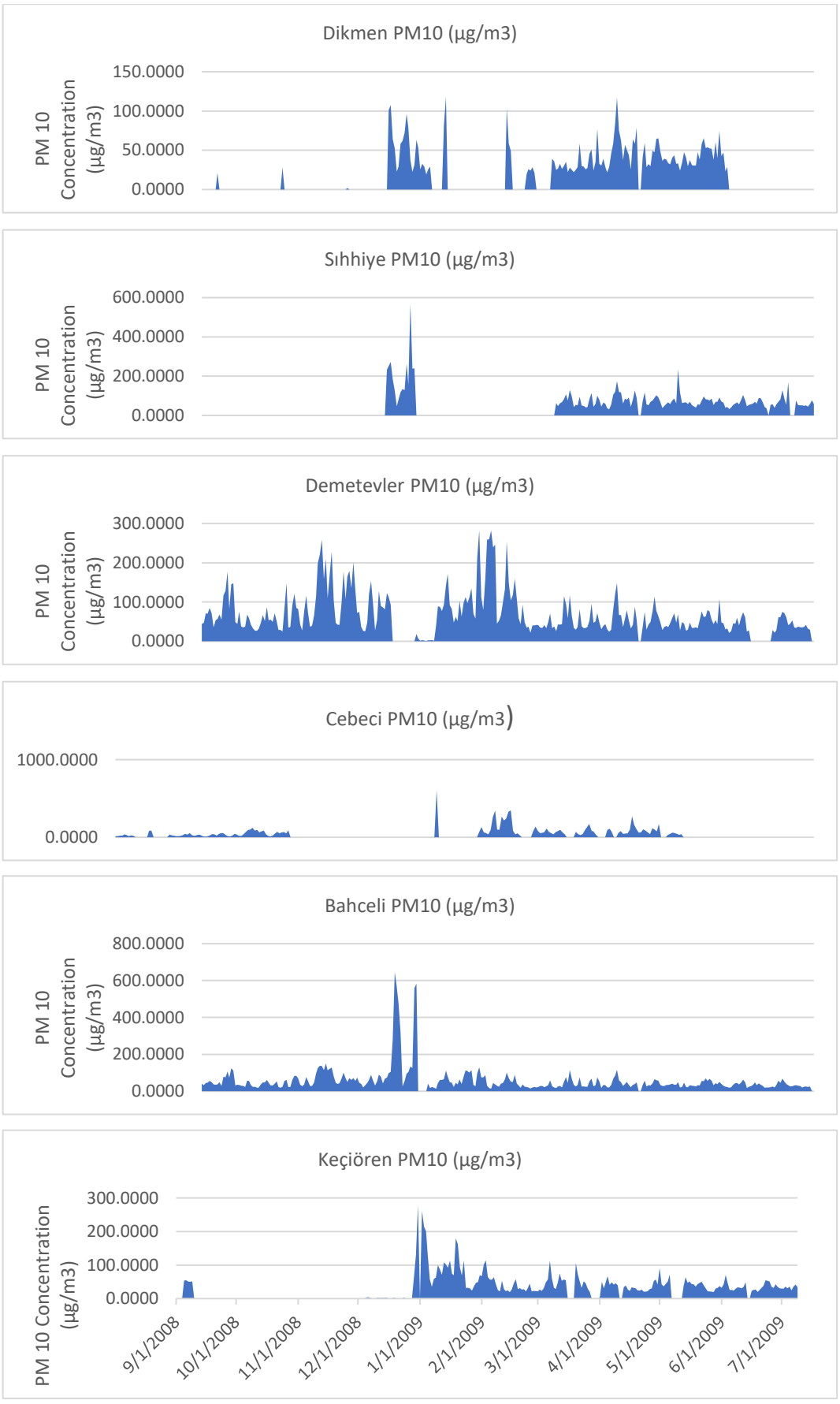

Figure 4.32. Hourly variation of PM10 concentrations at regulatory monitoring stations at Ankara.

The event of March is a typical case of Saharan dust transport affecting Ankara. On 3 March south-southeast direction wind resulted in low particulate matter concentrations around Ankara. But at 4 March persistent north-northeastern flows are observed. Thus, a Saharan plume expanded along with Turkey as seen in figure 4.33 which resulted in high particulate matter concentrations to be observed. Long-range dust transport is also observed in the back-trajectory figures. Greenline which shows the transport at 1500m in figures 4.36 shows the long-range transport on April episode from Libya to Turkey, Ankara. The subsequent particulate scavenging by rainfall resulted in decreases in particulate matter concentrations. This change in concentrations is seen better in figure 4.34 and 4.35. Highest particulate matter is observed in Cebeci station and the lowest concentration is observed in Kecioren station

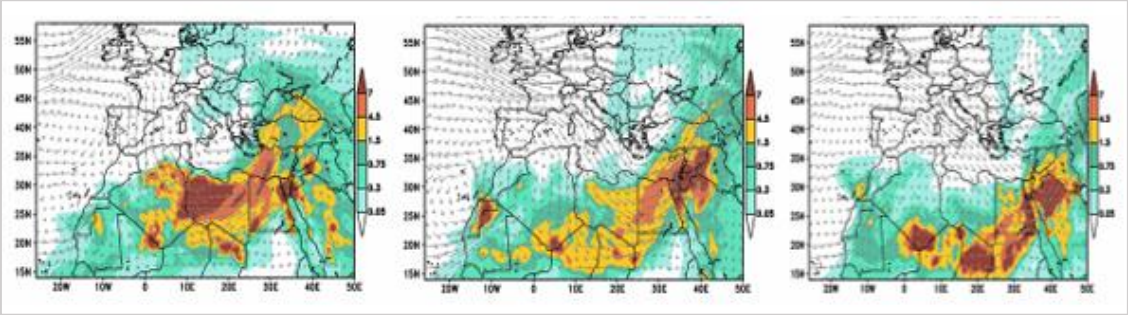

Figure 4.33. Sample dust episode for March dust event.

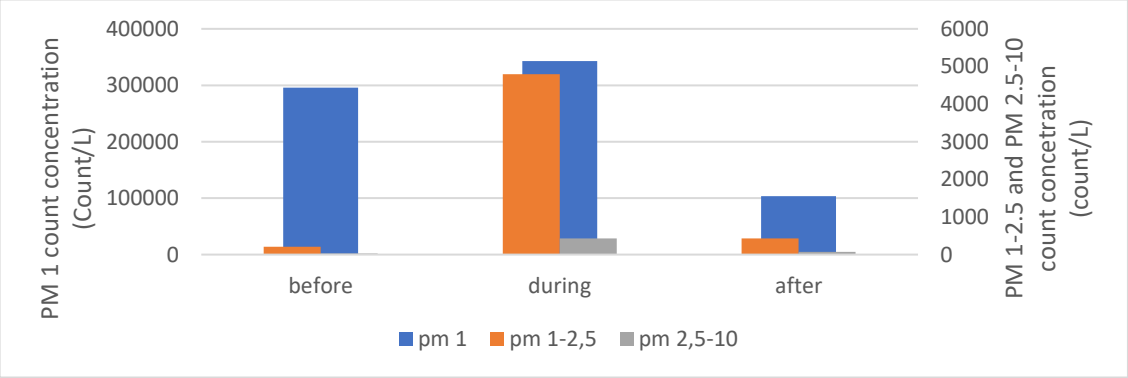

Figure 4.34. Change in the particle count concentration before, during and after March dust episode.

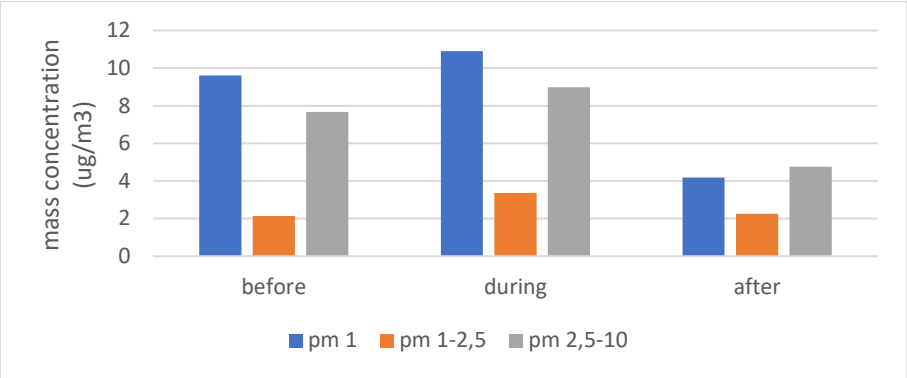

Figure 4.35. Change in the particle mass concentration before, during and after March dust episode.

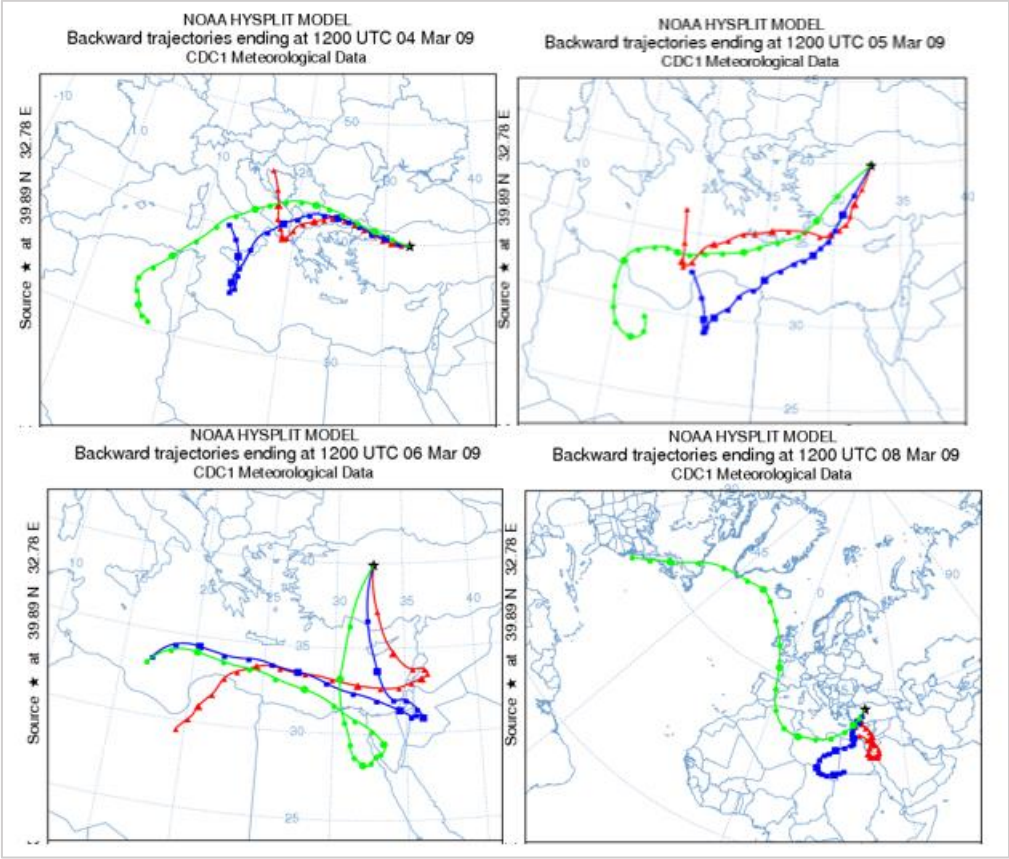

Figure 4.36. Back trajectory figures for the dust episode March.

Another interesting example that is showing the effect of Saharan dust episodes on the particulate matter concentration can be given as the episode found in April (figure 37, 38, 39, and 40). It is seen that stations are not affected by dust episodes in the same way. In Sihhiye station, the highest PM 10 concentration and in Dikmen station the lowest concentration is observed. Where in the previous example highest was Cebeci station and the lowest was Kecioren station. This shows the influence of the plume affecting different stations. As described in the previous dust episode, a sharp decrease in particulate matter concentration is observed with the rain on the April dust event.

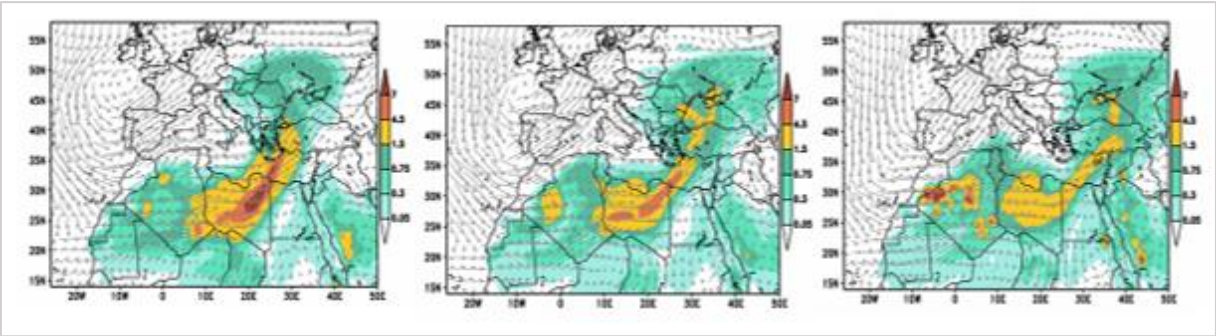

Figure 4.37. Sample dust episode.

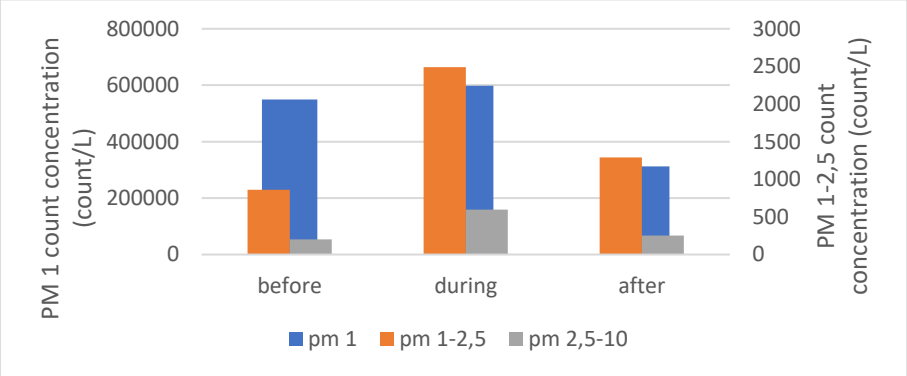

Figure 4.38. Change in the particle count concentration before, during and after April dust episode.

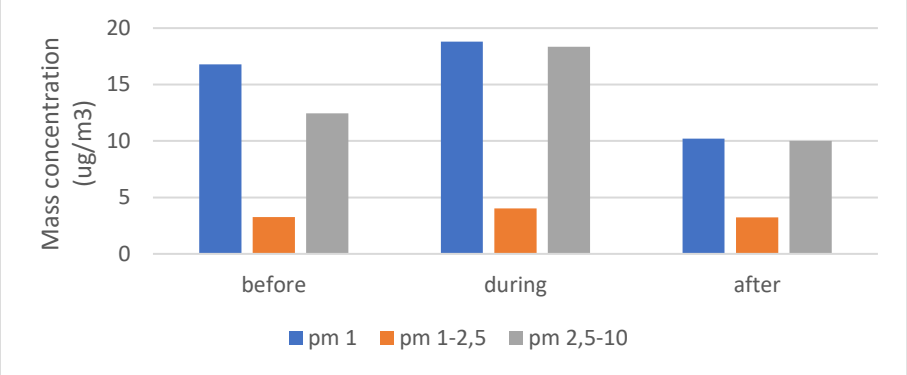

Figure 4.39. Change in the particle mass and count concentration before, during and after April dust episode.

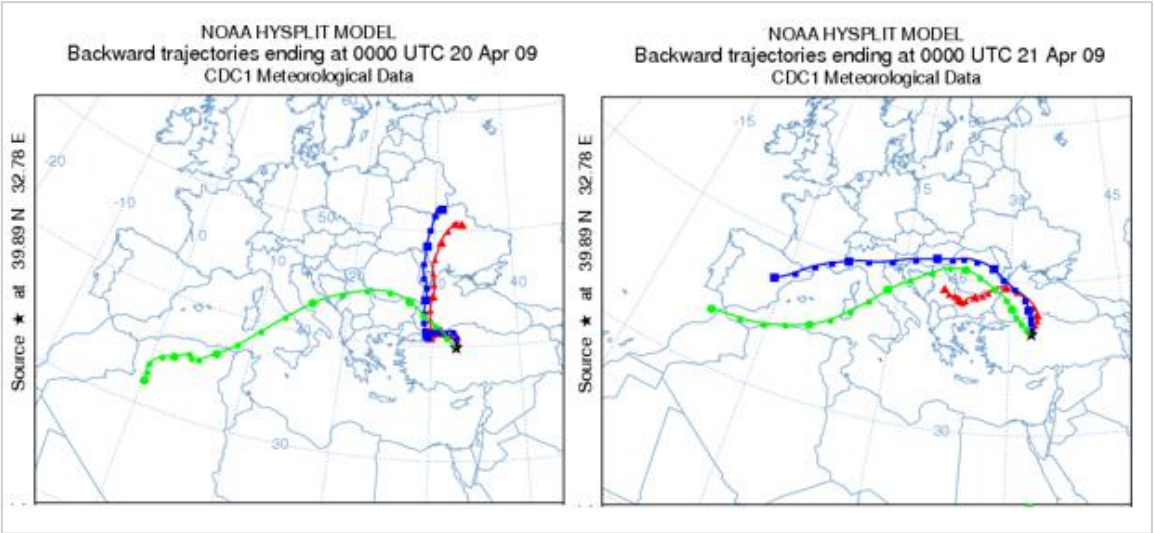

Figure 4.40. Back trajectory figures for the April dust episode.

In dust episodes, only on PM concentration should be increased. But in some determined dust episodes, it is seen that SO_2 concentration also increases. This increase of both particulate matter and SO_2 concentration means there is an anthropogenic episode during that time. In order to determine anthropogenic episodes, hourly SO_2 concentration data is collected from six stations mentioned before table 4.13. Collected data is divided into four seasonal groups which are autumn (Sept, Oct, Nov), winter (Dec, Jan, Feb), spring (Mar, Apr, May) and summer (June, July, Aug). For each season hourly SO_2 concentration data is sorted from largest to smallest and the SO_2 values that fit the 75th percentile of data set is determined as candidate anthropogenic episode. There is not a specific rule for the episodic concentration selection. But generally, three methods are used which are:

- Choosing an air quality limit concentration as a threshold value for episodes (Muir et al., 2006)
- Selecting a threshold level by using personal experience (Amodio et al., 2008)
- Recognizing the upper 25% of the data as an episode when data is sorted with decreasing order. (Karaca et al., 2005)

In this study, the last approach is used which is found to be an effective method to determine episodes (Karaca et al., 2009)

Data is divided into seasons because concentrations show different statistical characteristics with different seasons. Winter concentration is found to be nearly two times higher than summer concentration which is expected because of the increased household heating in winter. If the data is

not divided into seasons, because of the high concentration during winter, most of the episodes would be observed during winter. By dividing data, we get a chance to search seasonal anthropogenic episodes. Episode length is selected as 10 hours or higher as in the dust episode determination process. Episode candidates lower than 10-hour length is removed from the candidate data. This removal is done in order to minimize the effect of daily increase and decrease in SO2 concentrations between rush hours (08.00-17.00) and the effects of meteorology. Finally, the remaining data is selected as anthropogenic episode data.

Table 4.13. Summary statistics of hourly SO_2 mass concentrations (in ug/m^3) of each station.

	Bahcelievler SO_2	Cebeci SO_2	Demetevler SO_2	Dikmen SO_2	Kecioren SO_2	Sihhiye SO_2
count	6879	2821	6401	6533	6932	4568
blank count	393	4451	871	739	340	2704
mean	26,150	10,749	14,060	10,988	9,326	12,851
std error of mean	0,332	0,194	0,464	0,178	0,109	0,187
median	16,000	8,000	6,000	7,000	7,000	10,000
mode	9,000	6,000	3,000	5,000	7,000	0,000
std deviation	27,529	10,286	37,092	14,358	9,048	12,649
variance	757,864	105,795	1375,814	206,140	81,871	159,985
skewness	2,677	3,199	6,182	4,539	3,688	2,547
kurtosis	11,046	18,047	45,192	30,401	26,448	11,341
maximum	307	108	504	197	139	136

As a result of this determination of dust and anthropogenic episode process, 19 dust episodes and 15 anthropogenic episodes are found to fit the previously mentioned criteria.

In order to see if there are anthropogenic episodes during dust episodes, line graphs of PM 10 and SO_2 concentrations are given in figure 4.41. In both figures and data set many anthropogenic episodes are observed during dust episodes.

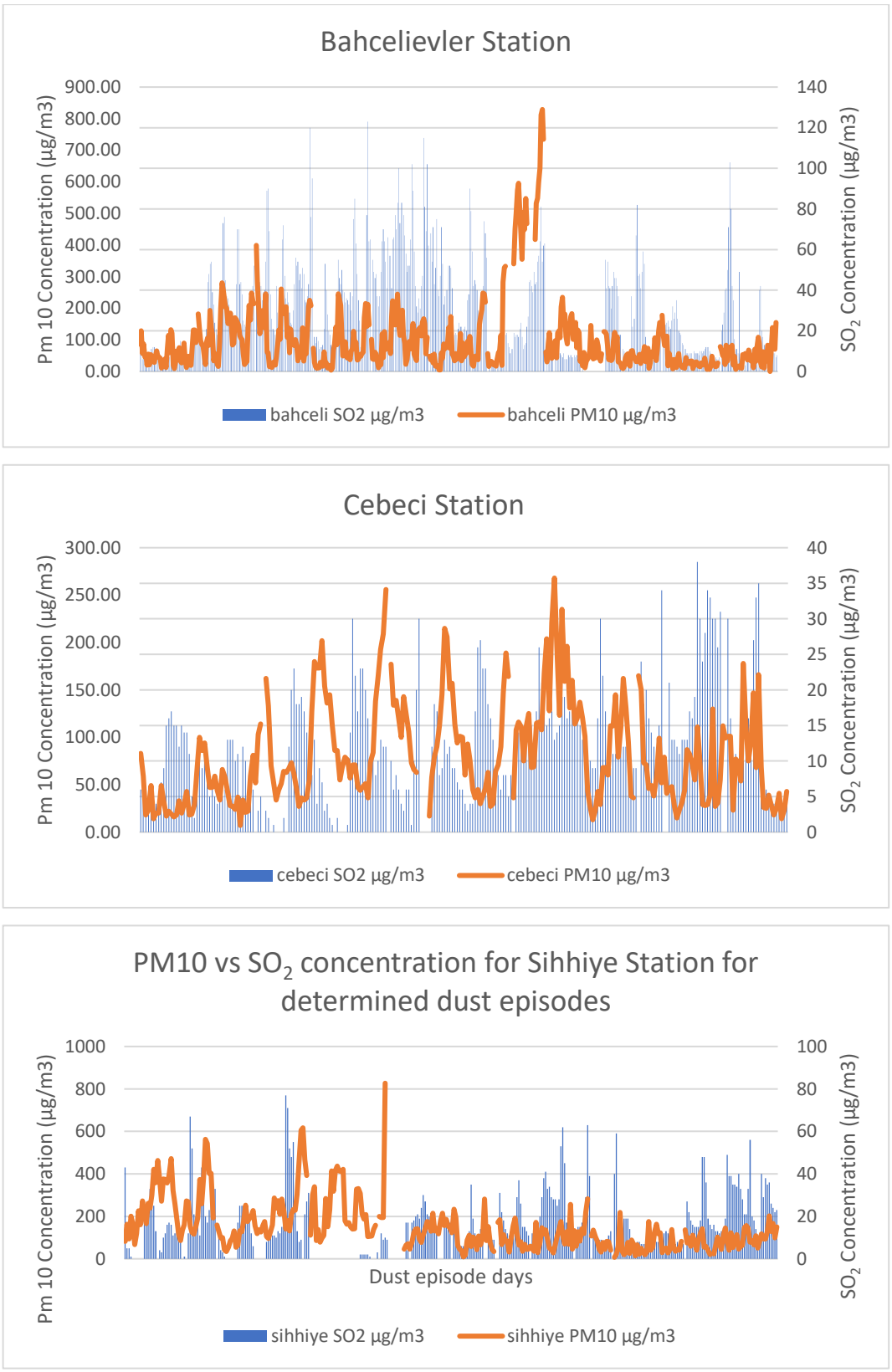

Figure 4.41. SO_2 and PM 10 concentration distribution for three stations during dust episode days.

In table 4.14 change of particulate matter and SO_2 concentrations with anthropogenic and dust episodes is given. In both anthropogenic and dust episodes, particulate matter concentration increases considerably in all stations, but SO_2 concentration does not change significantly for dust episodes where nearly 2-3 times higher concentrations are observed in anthropogenic episodes which are expected.

Table 4.14. Particulate matter and SO_2 concentrations ($\mu g/m^3$) during anthropogenic and dust episodes.

	Bahcelievler PM10	Cebeci PM10	Demetevler PM10	Dikmen PM10	Keçiören PM10	Sıhhiye PM10
Both anthropogenic and dust episode	90,39	90,95	111,63	70,57	49,43	123,03
Anthropogenic episode	79,10	62,87	123,45	59,32	67,95	94,70
Dust episode	69,51	90,81	86,59	50,15	48,00	93,66
No episode	44,28	51,45	57,91	36,93	46,03	71,14

	Bahcelievler SO_2	Cebeci SO_2	Demetevler SO_2	Dikmen SO_2	Kecioren SO_2	Sihhiye SO_2
Both anthropogenic and dust episode	53,10	29,04	38,28	25,20	16,61	28,85
Anthropogenic episode	55,67	10,92	29,36	26,34	17,51	19,08
Dust episode	26,92	14,12	11,43	11,45	10,89	16,32
No episode	20,89	8,18	11,64	8,03	7,37	9,67

In order to see the particle count concentration change with dust episodes, identified dust episode days of station data are used to find a relation between particle count concentration data that we have collected for a 12-month period with previously mentioned equipment. Dust episode days are highlighted and two days before and two days after the dust episode data is given. In figure 4.42, change of particle count concentrations before, during, and after dust episodes are given for different diameters. It is seen that during dust episodes, particle count concentration does not change for particles with a diameter of less than 0.65-0.70 µm. This shows that the particles moved with long-range transport are mostly particles bigger than 0.65-0.75 µm. During the dust episode, most of the particle sizes showed a significant concentration change before, during, and after the dust episode.

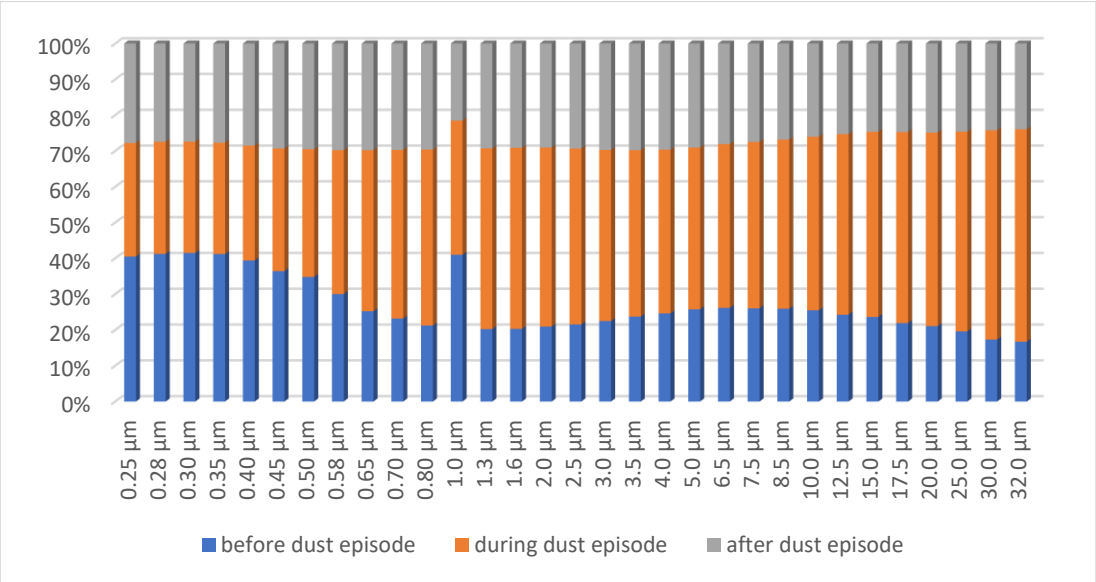

Figure 4.42. Change of particle count concentration before, during and after dust episodes.

In figure 4.43 and figure 4.44 variation of PM 1, PM 1-2.5, and PM 2.5-10 of both particle count and mass concentration with dust episode days. For PM 1 it is seen that mostly 1 day before dust episode concentration is higher than dust episode day's concentration but for PM 1-2.5 and PM 2.5-10 there is a significant concentration increase in dust episode days and concentration decreases again after dust episode which is seen also in the previous figure.

In table 4.15 and table 4.16, change of PM 1, PM 1-2.5, and PM 2.5-10 concentration with each dust episode when compared with the concentrations two days before the dust episode. To better understand the changes of particle number and mass concentration with dust events figures 4.45 and 4.46 are generated from the tables 4.15 and 4.16. In some episodes, some negative percentages are found and when data is analyzed in detail, it is found that the main reason for the percentage to be negative is the occurrence of anthropogenic episodes before dust episodes and so calculated concentration of two days before dust episode days is higher than dust episode days.

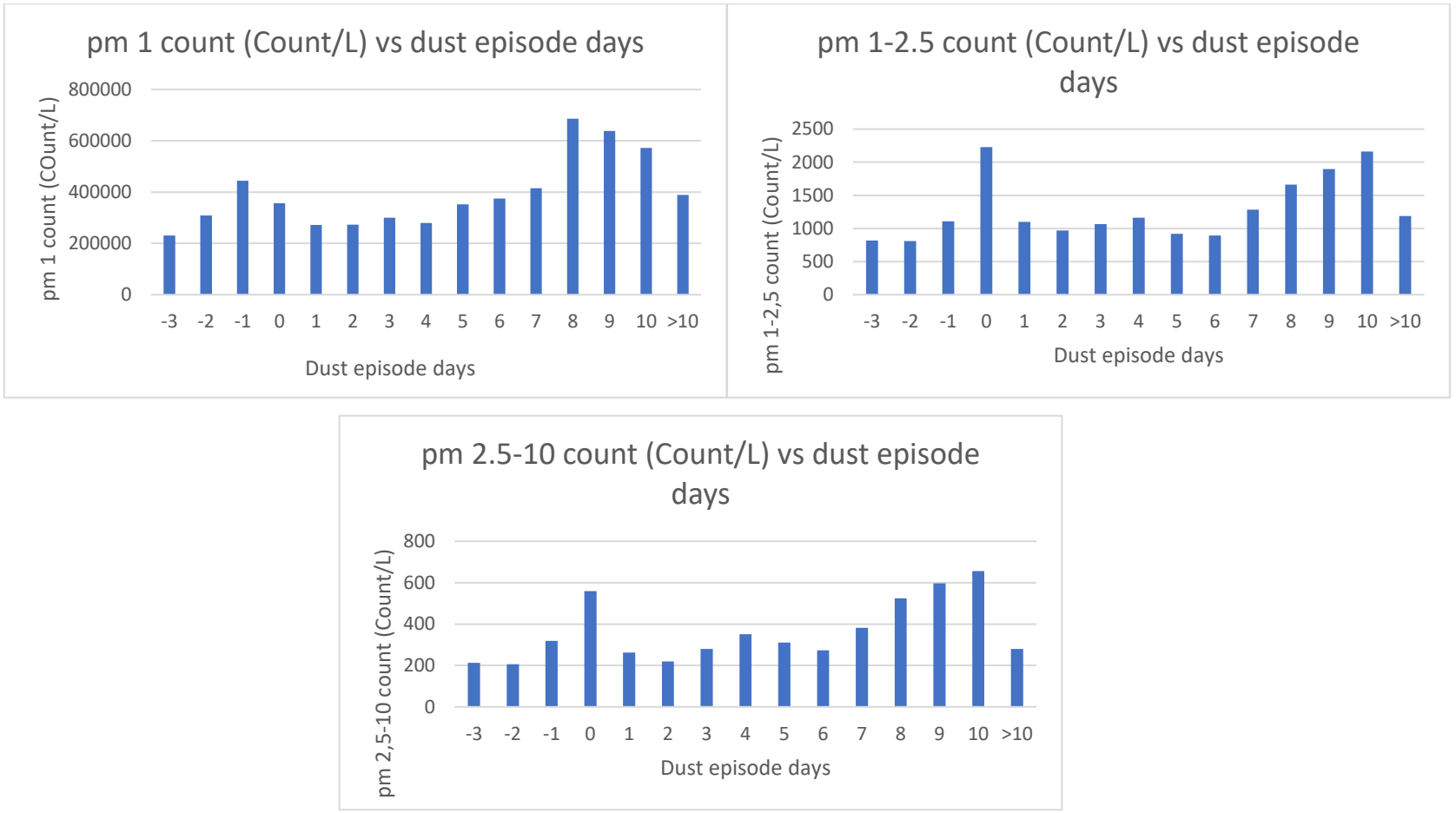

Figure 4.43. Variation of PM 1, PM 1-2.5 and PM 2.5-10 for particle count concentration with dust episode days.

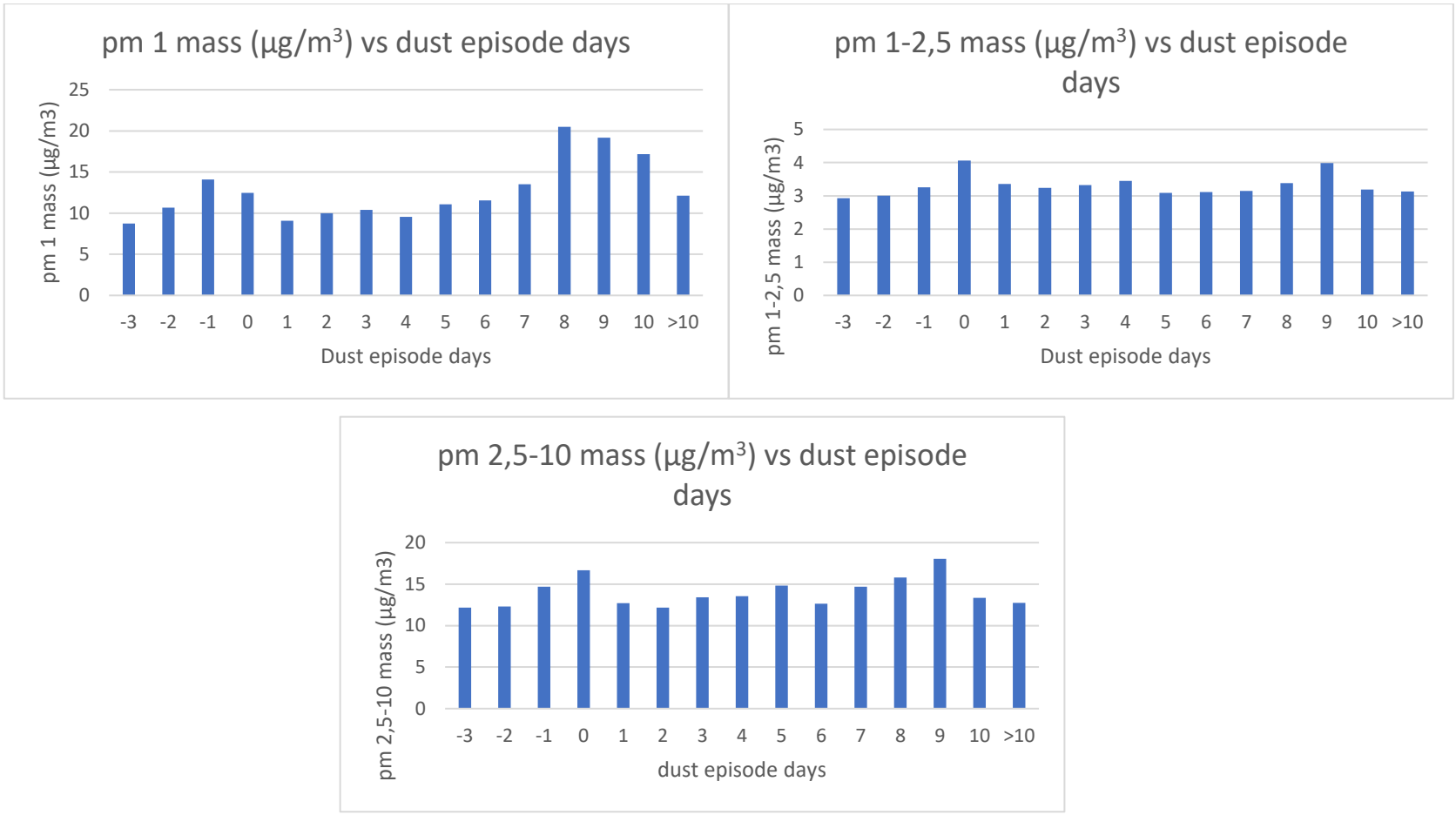

Figure 4.44. Variation of PM 1, PM 1-2.5 and PM 2.5-10 for particle mass concentration with dust episode day.

Table 4.15. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 number concentration during the dust episode compared with 2 days before concentrations.

	Average dust episode number concentration (Count/L)	The percentage increase of PM1 number concentration during the dust episode compared with 2 days before (%)	The percentage increase of PM1-2,5 number concentration during the dust episode compared with 2 days before (%)	The percentage increase of PM2,5-10 number concentration during the dust episode compared with 2 days before (%)
dust episode 1	383981,92	89,80	121,49	121,18
dust episode 2	258939,95	-26,88	283,39	206,74
dust episode 3	330801,46	-20,33	8,23	-5,36
dust episode 4	945860,12	42,49	207,51	194,30
dust episode 5	368467,41	160,24	452,79	567,72
dust episode 6	843201,12	-25,88	-19,25	-12,10
dust episode 7	1076045,28	130,53	161,81	171,61
dust episode 8	1039739,80	81,87	-83,38	374,46
dust episode 9	1343877,92	-25,30	-48,11	-59,96
dust episode 10	982980,54	-40,04	1,74	-8,13
dust episode 11	365597,35	-65,84	46,01	-13,89
dust episode 12	308301,83	-5,66	1180,80	1201,05
dust episode 13	568793,69	101,51	522,80	513,11
dust episode 14	473475,78	12,51	77,48	53,90
dust episode 15	591920,90	35,66	63,23	45,48
dust episode 16	325170,30	10,19	-3,56	33,83
dust episode 17	283338,99	-10,44	215,29	157,02
dust episode 18	200317,66	69,93	-50,57	98,99
dust episode 19	223815,50	5,88	6,87	42,65

Table 4.16. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 mass concentration during the dust episode compared with 2 days before concentrations.

	Average dust episode mass concentration (μg/m^3)	The percentage increase of PM1 mass concentration during the dust episode compared with 2 days before (%)	The percentage increase of PM1-2,5 mass concentration during the dust episode compared with 2 days before (%)	The percentage increase of PM2,5-10 mass concentration during the dust episode compared with 2 days before (%)
dust episode 1	39,28	60,31	20,09	16,58
dust episode 2	27,0	-20,89	26,30	6,31
dust episode 3	28,11	-17,84	14,32	-6,35
dust episode 4	51,41	41,41	31,04	48,71
dust episode 5	25,15	136,06	14,52	165,45
dust episode 6	45,11	-23,42	-0,41	-2,01
dust episode 7	47,62	115,25	285,70	72,93
dust episode 8	48,97	70,81	22,29	18,18
dust episode 9	63,89	-24,46	8,68	-15,80
dust episode 10	48,94	-38,66	-17,67	-29,01
dust episode 11	26,38	-62,68	-20,82	-48,60
dust episode 12	21,69	-4,16	63,11	14,70
dust episode 13	37,04	97,19	79,38	201,04
dust episode 14	32,95	13,55	13,73	34,84
dust episode 15	37,18	30,94	48,70	19,99
dust episode 16	27,13	10,70	10,68	20,80
dust episode 17	36,22	-5,99	22,23	12,26
dust episode 18	29,88	47,52	27,89	35,96
dust episode 19	33,09	10,26	15,61	26,74

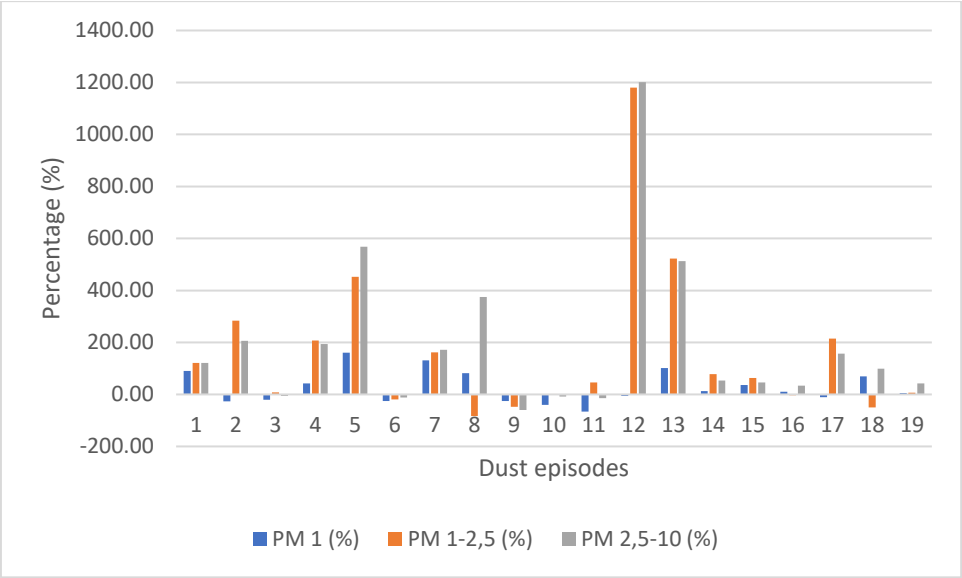

Figure 4.45. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 number concentration during the dust episode compared with 2 days before concentration.

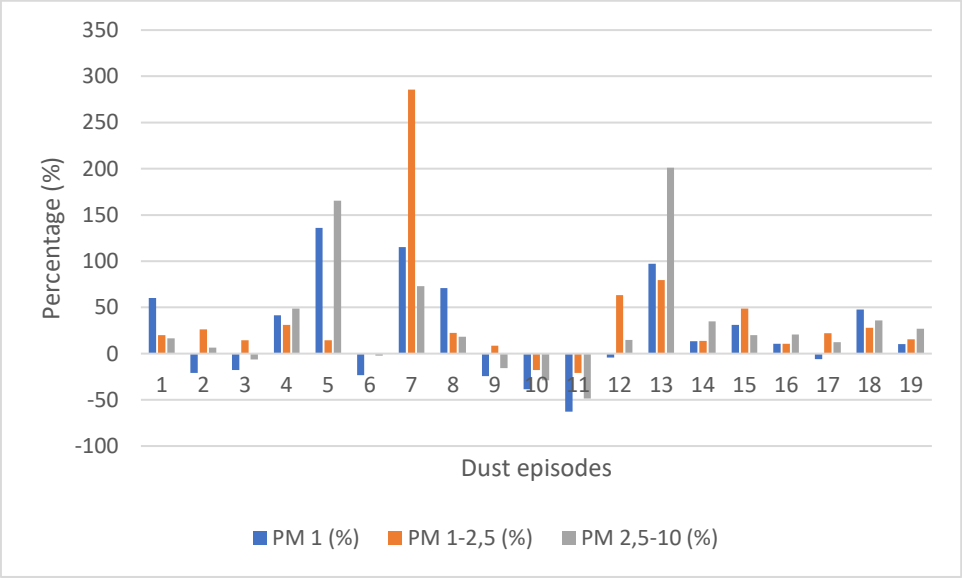

Figure 4.46. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 mass concentration during the dust episode compared with 2 days before concentration.

4.6. Variation of particle size distribution with anthropogenic episodes

Even short-term exposure to SO_2 may harm the human respiratory system. Children and people with asthma are much more sensitive to the effects of SO_2. Emissions that lead to high SO_2 concentration in the atmosphere also lead to SOx formation. We observe these particles as particulate matter and as a result, SOx contribute to particulate matter pollution and, the resulting health and

environmental effects are observed. Fine particles penetrate deeply into the lungs and create respiratory-related health problems, also these particles damage plant foliage and decrease plant growth (EPA, 2020).

Change of particle count concentration before, during, and after SO_2 (anthropogenic) episodes are given in figure 4.47. It is seen that anthropogenic episode is more affective on fine particles and decreases with the increasing particle diameter. Concentrations of fine particles increase with the episode and decrease after the episode which we saw the opposite in dust episodes, there was an increase in concentration for particles 0.65-0.70 µm but smaller particles are not affected that much. As a result, it can be said that with anthropogenic episodes, mostly fine particles are transported.

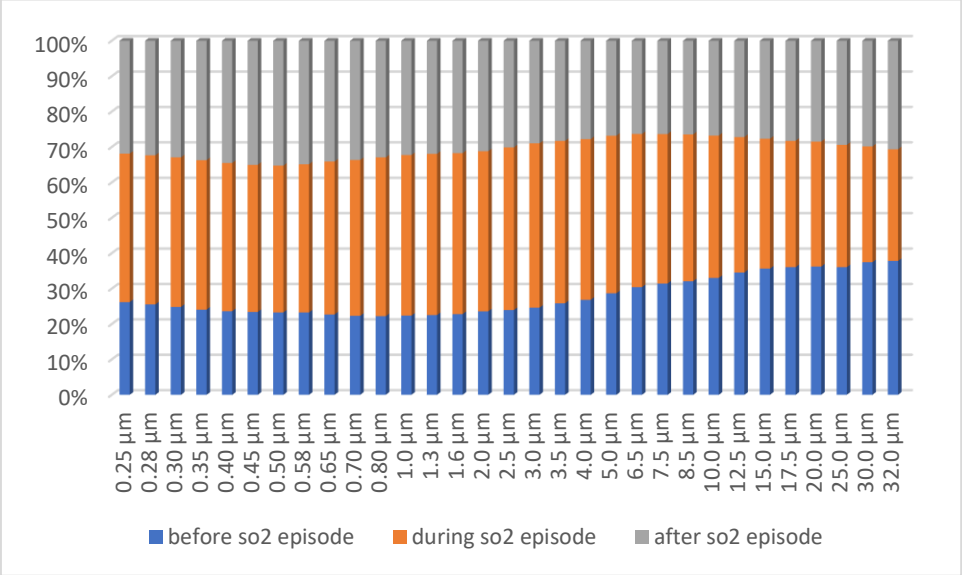

Figure 4.47. Change of particle count concentration before, during and after SO_2 (anthropogenic) episodes.

In figure 4.48 and 4.49 particle count and mass concentration change with two days before, during and 10 days after anthropogenic (SO_2) episodes are given. The effect of the anthropogenic episode is seen well in these figures. There is a sharp increase in the episode days and a sharp decrease after the episode for particle count concentration of PM 1, PM 1-2.5 and PM 10. Mainly 5 days after the episode, concentration reaches its minimum and starts to increase. But for the mass concentration data, a significant increase and decrease before and after anthropogenic transport are observed only on PM 1 and PM 1-2.5 mass concentration data. For PM 2.5-10 concentration, the anthropogenic episode does not have as much effect as for PM 1 and PM 1-2.5 concentration. This shows that

anthropogenic episode, which is previously mentioned, is caused by household heating mostly that produces fine particles and as a result mostly affective on fine particle concentrations.

In table 4.17 and table 4.18, change of PM 1, PM 1-2.5, and PM 2.5-10 concentration with each anthropogenic episode when compared with the concentrations two days before anthropogenic episode. To better understand the changes of particle number and mass concentration with anthropogenic events figures 4.50 and 4.51 are generated from the tables 4.17 and 4.18. In some episodes, some negative percentages are found this may be due to miscalculated data from the data collection stations or other sources that are contributing to a high concentration of particulate matter.

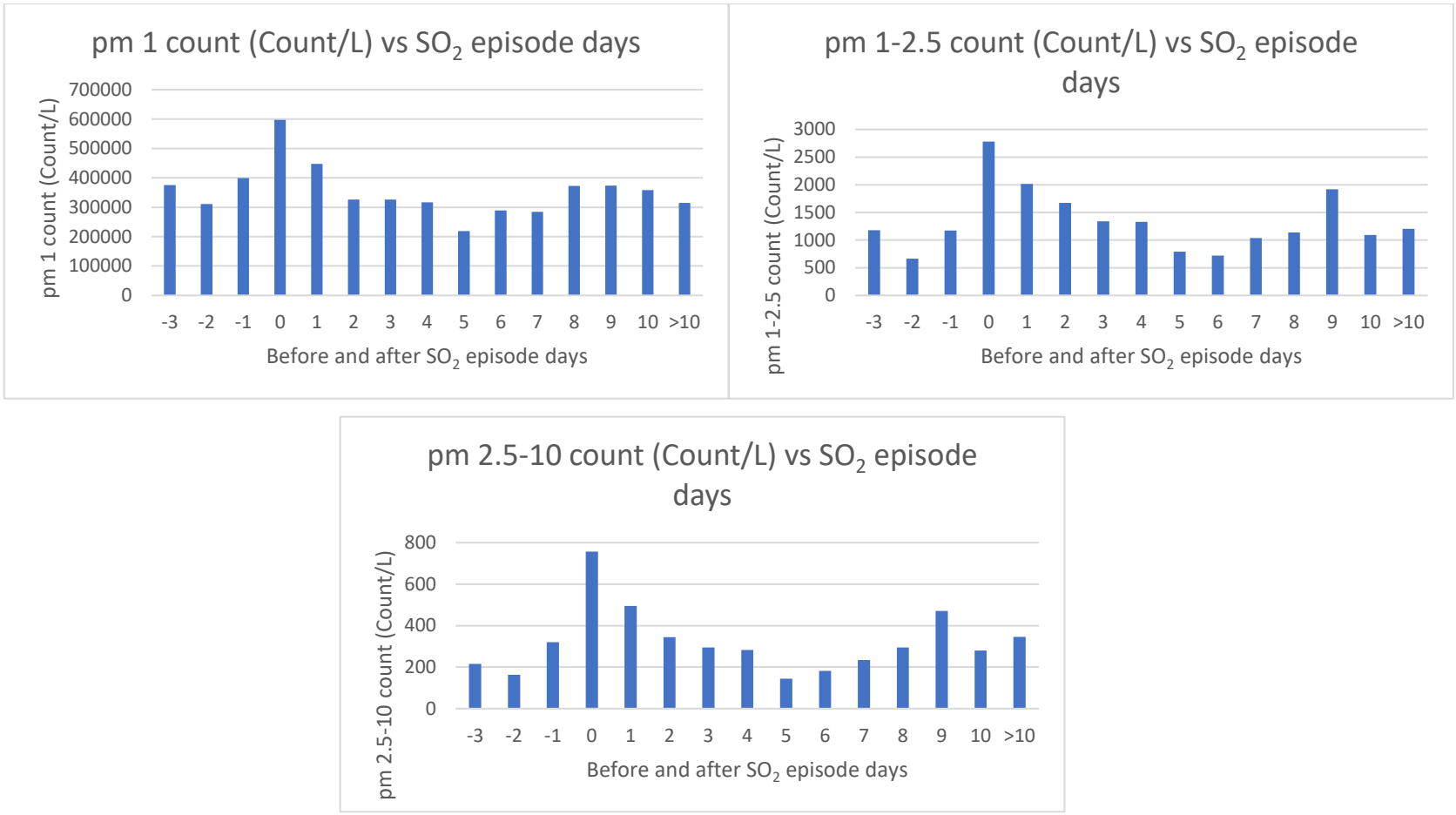

Figure 4.48. Variation of PM 1, PM 1-2.5 and PM 2.5-10 for particle count concentration with anthropogenic episode days.

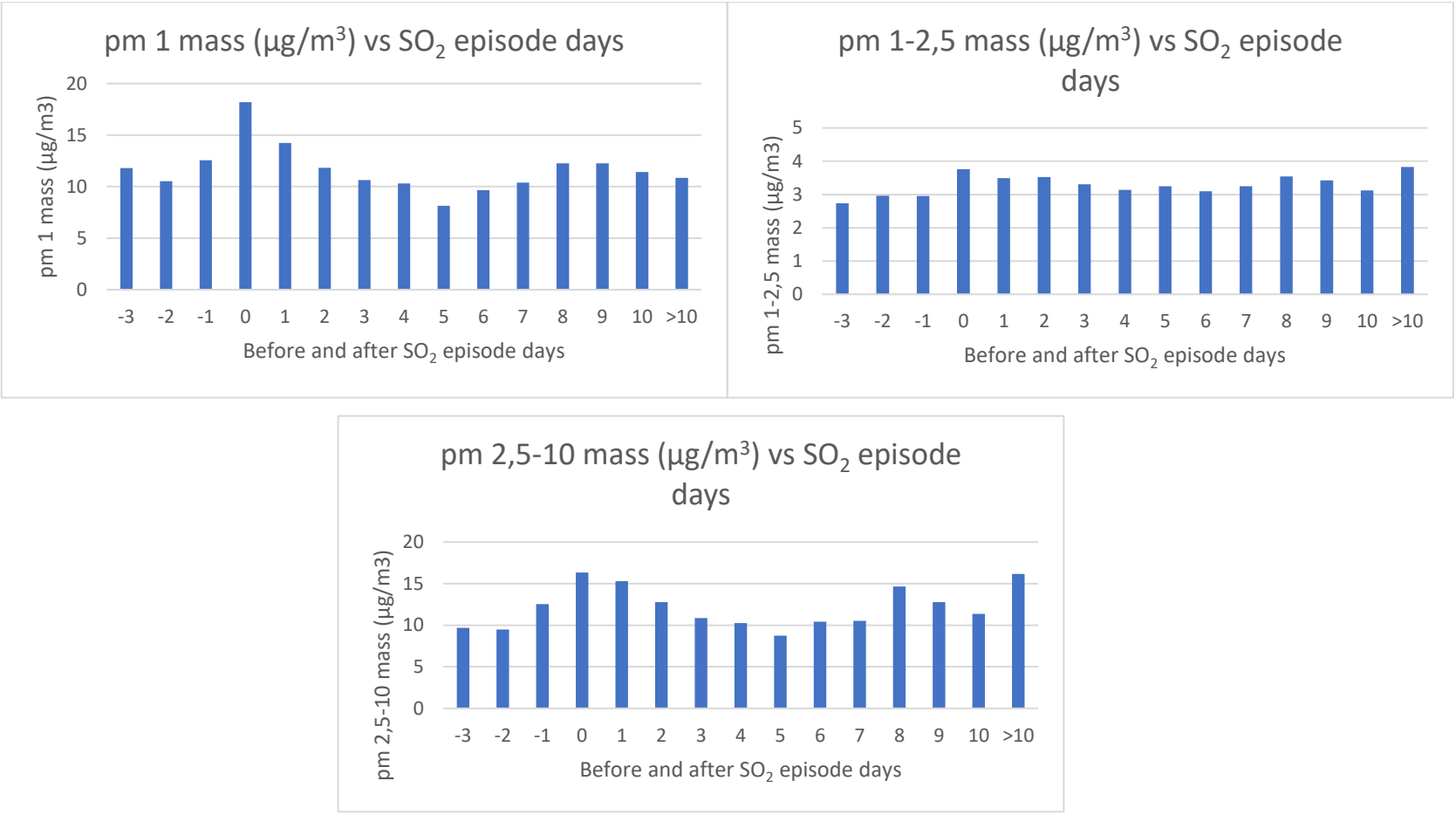

Figure 4.49. Variation of PM 1, PM 1-2.5 and PM 2.5-10 for particle mass concentration with anthropogenic episode day.

Table 4.17. The percentage increase of PM 1, PM 1-2.5 and PM 2.5-10 particle count concentration during the anthropogenic episode compared with 2 days before concentration.

	Average Anthropogenic episode number concentration (Count/L)	The percentage increase of PM1 number concentration during the anthropogenic episode compared with 2 days before (%)	The percentage increase of PM1-2,5 number concentration during the anthropogenic episode compared with 2 days before (%)	The percentage increase of PM2,5-10 number concentration during the anthropogenic episode compared with 2 days before (%)
Anthropogenic episode 1	643388,54	67,87	54,03	52,02
Anthropogenic episode 2	707617,98	-8,64	3,97	25,86
Anthropogenic episode 3	1461233,31	66,89	-32,20	-53,94
Anthropogenic episode 4	1137250,57	208,51	161,91	128,27
Anthropogenic episode 5	1623140,91	106,86	287,12	245,31
Anthropogenic episode 6	1720091,59	84,57	146,89	184,39
Anthropogenic episode 7	555036,28	50,01	304,32	195,17
Anthropogenic episode 8	982005,24	142,22	286,02	577,45
Anthropogenic episode 9	346290,55	16,75	149,79	223,25
Anthropogenic episode 10	406806,26	-26,01	139,32	202,44
Anthropogenic episode 11	568793,69	101,51	522,79	513,10
Anthropogenic episode 12	198010,08	48,81	89,54	54,93
Anthropogenic episode 13	519364,77	235,51	210,91	234,10
Anthropogenic episode 14	202894,41	145,56	84,69	157,60
Anthropogenic episode 15	438374,86	26,39	151,21	190,70

Table 4.18. The percentage increase of PM 1, PM 1-2.5 and PM 2.5-10 mass concentration during the anthropogenic episode compared with 2 days before concentration.

	Average Anthropogenic episode mass concentration ($\mu g/m^3$)	The percentage increase of PM1 mass concentration during the anthropogenic episode compared with 2 days before (%)	The percentage increase of PM1-2,5 mass concentration during the anthropogenic episode compared with 2 days before (%)	The percentage increase of PM2,5-10 mass concentration during the anthropogenic episode compared with 2 days before (%)
Anthropogenic episode 1	37,50	55,19	2,36	10,38
Anthropogenic episode 2	42,53	-7,66	-4,76	-5,17
Anthropogenic episode 3	64,85	59,54	17,26	12,47
Anthropogenic episode 4	53,09	183,52	125,11	131,41
Anthropogenic episode 5	74,91	101,94	178,87	115,58
Anthropogenic episode 6	79,24	81,92	242,83	112,35
Anthropogenic episode 7	35,22	43,74	1,89	29,06
Anthropogenic episode 8	47,10	131,32	39,47	129,31
Anthropogenic episode 9	20,03	12,82	-6,71	-5,92
Anthropogenic episode 10	22,88	-21,71	-8,37	14,86
Anthropogenic episode 11	37,04	97,19	79,38	201,03
Anthropogenic episode 12	33,85	27,32	22,30	8,80
Anthropogenic episode 13	28,79	181,14	73,88	113,62
Anthropogenic episode 14	15,84	77,97	35,26	-5,68
Anthropogenic episode 15	32,58	24,13	-2,44	17,25

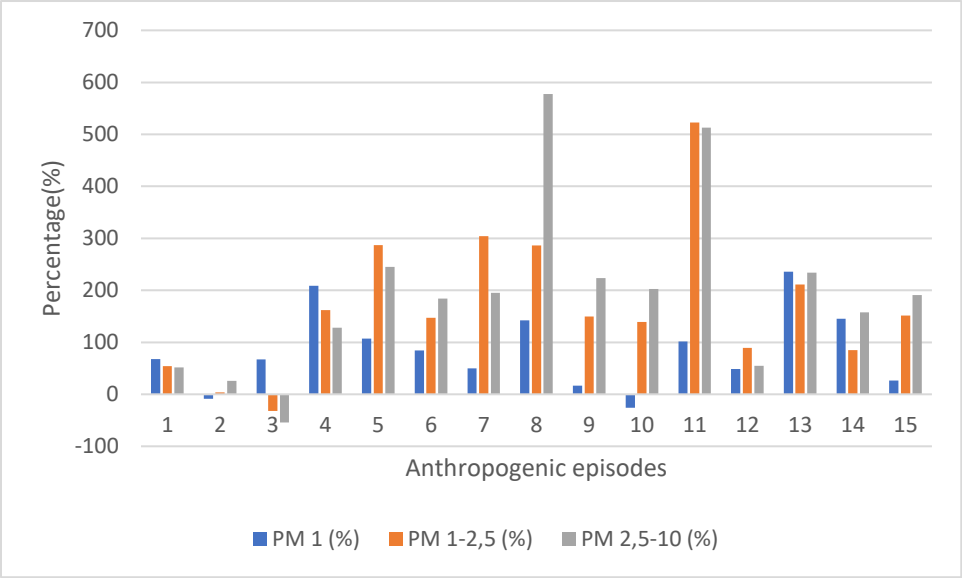

Figure 4.50. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 number concentration during the anthropogenic episode compared with 2 days before concentration.

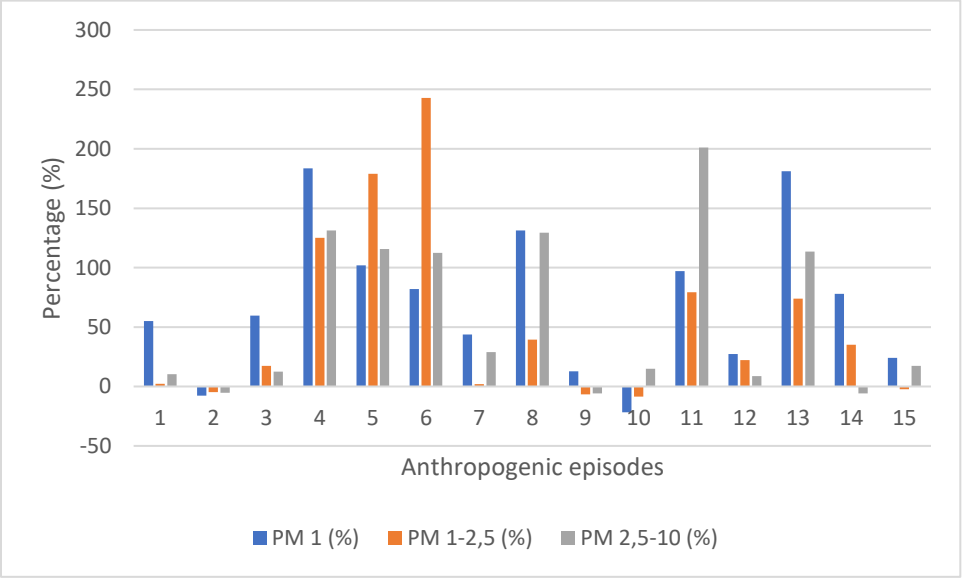

Figure 4.51. The percentage increase of PM 1- PM 1-2.5 and PM 2.5-10 mass concentration during the anthropogenic episode compared with 2 days before concentration.

5. CONCLUSIONS

In this study, particulate matter count concentrations are determined by using a particle counter from the GRIMM company named Ambient Aerosol Monitor with Integrated Sampling Pipe Heater Model 265. Data is collected for every minute of a 12-month period, in 31 different sizes form 0.25µm to 32 µm. By the same time a stacked filter unit is used at the same place as the particle counter, in order to determine mass concentrations for PM10 and PM2.5. Particle counter device determines particle count concentration and by using this data it calculates the mass concentration with its own model and regression analysis. The GRIMM company provided mass concentration data for three months.

At the same time with particle count data collection, particle mass concentration data is collected by using a stack filter unit. By using the stack filter concentration data and the mass concentration data provided by the GRIMM company, conversion equation from the particle count concentration data to particle mass concentration data is modeled by using multiple regression. In the conversion equation, environmental factors such as temperature, humidity, and pressure are found to be relevant and included in the equation.

With particle count concentration and mass concentration data, effects of environmental factors such as wind speed, humidity, wind direction, temperature, solar flux are determined. It is seen that effect of wind speed changes for different particle sizes. This change showed us the effect of resuspension, transport of particles from other areas. For example, PM1 decreases with the increasing wind speed but for larger particles increasing wind speed increases the particle concentration. N-NE (north-northeast) wind direction is found to be the most dominant wind direction that affects both particle count and mass concentration data which may be due to the increased particulate matter concentrations as a result of the dust transport from Saharan desert or pollutant sources from the dominant wind direction. As a result of the decreased household heating and combustion, particulate matter concentrations decreased with the increasing temperature. An increase is observed with increasing temperature for larger sized particles which showed us the effect of photochemical reactions between precursors. With the increasing humidity, particle count and mass concentrations increased for PM 1. Different distribution is observed for larger particles since they grow too heavy and with dry deposition, particles are removed so a decrease is observed with higher humidity.

Daily rainfall data is collected from the Etimesgut air pollution control station. With the rain data, particle number and mass concentrations are compared from -2 day before a rain event to 10 days after. It is seen that on rainy days there is a significant concentration reduction due to wet deposition and removal of particles from the atmosphere. Also, it is seen that for particles smaller than 1 μm, there is not much difference between rainy and non-rainy days. But larger particles are seen to be lower on rainy days which shows that coarse particles are removed more effectively by rain.

Diurnal, weekday-weekend, monthly variation of particle mass and count concentrations are compared. In a diurnal variation of particle mass and count concentration, the highest concentrations are observed during rush hours. In the weekday-weekend variation, generally, there is not much difference between weekday and weekend in the count concentrations except PM 1-2.5 and PM 2.5-10 range but mass concentrations are mainly found to be higher on weekdays. Monthly and seasonal averages are calculated to see the change of particulate mass and number concentrations with different months. In winter there is an increase in both mass and number concentrations as expected. Also, it is seen that since household heating produces small particles, at winter increase at the submicron particles is observed.

In order to see the effect of dust and anthropogenic events on mass and number concentration data, both PM 10 and SO_2 concentrations are collected from six sampling stations. By comparing the concentrations with different models dust and anthropogenic episodes are determined.

19 dust episodes determined during the sampling time. In dust episodes, it is seen that the most significant increase is observed for PM 1-2.5 and decreases after the episode. This decrease lasts generally a week the increase is mainly observed in the count concentration but since these particles do not have much effect on mass concentration a significant increase is not observed for mass concentration. It is also seen that after dust episodes PM 1 count and mass concentration decrease significantly but there is not much difference between before and during dust episode concentrations. A sharp increase and decrease before and after dust episode observed for PM 10 mass and count concentration data. Sharp decreases after the episode maybe as a result of the washout of particles with rain.

In some dust episodes, there is also an increase in the SO_2 concentrations which means anthropogenic episodes may occur. The most significant increase in PM10 concentrations in all stations seen when there are both anthropogenic and dust episodes. Also, SO_2 concentration is nearly 2-3 times higher than the concentrations observed during dust episodes.

Anthropogenic episodes mainly affect particles smaller than 2.5 μm. Concentrations of fine particles increase with episodes and decrease afterward. This decrease after the anthropogenic episode lasts generally 7 days after that the increase in the concentration is observed. When particle count and mass concentration data is compared with the concentrations two days before the episode, significant increases are observed for most of the cases.

5.1. Recommendations for future studies

Concentrations of pollutants emitted to the atmosphere as a result of the combustion and household heating process, which are PM 10 and SO_2 decreased significantly in the last 15 years due to the improved fuel quality and natural gas usage for households heating. However, the rate of natural gas usage should be increased more mainly in the squatter settlements and further fuel improvements should be searched in order to decrease pollutants emitted to the atmosphere.

In this study how particle count and mass concentration change with different environmental factors, different seasons, and how concentrations vary with dust and anthropogenic episodes are studied. PM 10 and SO_2 data is collected from the 6 sampling stations in Ankara but there was a lack of data in some stations which made the episode determination process more complex. Obviously, a more comprehensive measurement is needed.

There are many factors that determine the health and environmental effects of particulate matter, but the main property can be defined as the size of the particle. Small particles can reach the lungs and cause important health problems such as premature death, asthma, heart attacks. In TURKEY there is no emission regulation for PM 2.5. In U.S. and EU concentrations are determined 15 $\mu g/m^3$ and 25 $\mu g/m^3$ respectively. It should be regulated for TURKEY to determine a limit concentration for PM 2.5 in order to prevent negative health and environmental effects of particulate matter.

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