

**DETERMINATION OF THE NUMBER SIZE DISTRIBUTION OF
SUBMICRON ATMOSPHERIC AEROSOLS USING A
DIFFERENTIAL MOBILITY PARTICLE SIZER**

By

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**DISSERTATION SUBMITTED IN FULFILLMENT OF THE
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UNIVERSITY**

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MAY 2013

DECLARATION

I hereby declare that the thesis entitled "The Determination of the number size distribution of submicron atmospheric aerosols using a differential mobility particle sizer" herein submitted, is the result of my own investigation and research and has not been submitted in part or in full for any other degree or to any other University. Where other sources of information have been used, they have been acknowledged.

Candidate: M.D Mabaso, BSc, BSc (Hons)

Signature:

Date:

I hereby declare that this thesis is the candidate's original work. It is therefore submitted with my approval.

Supervisor: N.A. Kgabi, PhD

Signature:

Date:

DEDICATION

This work is dedicated to my son Junior and my family for their support and love.

ACKNOWLEDGEMENT

I would like to express my gratitude to all people who assisted me to complete this dissertation. I want to thank the Department of Physics staff members for giving me permission to commence this study, and to do the necessary experimental work using the departmental equipment in collaboration with Helsinki University.

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ABBREVIATIONS

VOCs	Volatile organic compounds
CCN	Cloud condensation nuclei
D_p	Diameter
D_a	Aerodynamic diameter
IPCC	The intergovernmental Panel on Climate Change
TSP	Total suspended particulate matter
PM10	Particulate matter less than 10 μ m in size,
PM2.5	Particulate matter less than 2.5 μ m diameter,
EPA	Environmental Protection Agency
DMPS	Differential Mobility Particle Sizer
DMA	Differential mobility analysis
Pt/cc	Particles per cubic centimeter
CPC	Condensation Particle Counter
LPM	Litres per minute
RH	Relative humidity
SO ₂	Sulphur Dioxide
O ₃	Ozone
NO _x	Nitrogen Oxides
CO	Carbon Monoxide
H ₂ SO ₄	Sulphuric Acid
TSI	Trust Science Innovation

ABSTRACT

Ultra-fine aerosol particles may contain excessive amounts of harmful substances that can be easily inhaled and eventually enter into the human blood stream. The objective of this study was to determine the number size distribution of aerosol particles in the size range of 0.01 – 0.8 μ m, and to investigate the existence of new particle formation and growth events in the Botsalano Game Reserve, which is a typical savannah environment.

The average daily particle number concentration ranged from 740 cm⁻³ to 10000 cm⁻³ and the particle growth was observed mainly within the range of 10 to 100nm. Seasonal variations in the total particle number concentrations were observed with high values in summer and low values in winter. It has also been observed that particle formation and growth events can be observed mainly during the day. The differential mobility particle sizer proved to be a relevant tool for determination of particle size distribution.

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CHAPTER 1

1.1. INTRODUCTION AND RATIONALE

The word aerosol was introduced more than 80 years ago as an analogy to the term hydrosol, a stable liquid suspension of solid particles (Hinds, 1999). Aerosols are defined as solid or liquid particles suspended in a gas, and they include a wide range of substances such as dust, fume, smoke, mist, fog, haze, and smog (Seinfeld and Pandis, 1998). Atmospheric aerosols are generally considered to be particles that range in size from a few nanometers (nm) to tens of micrometers (μm) in diameter.

Particles may be either directly emitted into the atmosphere or formed in the atmosphere by the oxidation of precursor gases such as sulphur dioxide, nitrogen oxides and volatile organic compounds (VOCs), where the resulting oxidation products nucleate to form new particles or condense on pre-existing ones. Particles formed through these two routes are referred to as primary and secondary particles respectively (Finlayson-Pitts and Pitts, 1999; Seinfeld and Pandis, 1998). Particles in the atmosphere arise from natural sources as well as anthropogenic activities (Seinfeld and Pandis, 1998).

Aerosols have the potential to significantly influence our entire planet through their role in heterogeneous chemistry in the troposphere and stratosphere (Finlayson-Pitts and Pitts, 2000), as well as their effect on the Earth's climate as they scatter sunlight and serve as condensation nuclei for cloud droplet formation (Charlson et al., 1992; Penner et al., 2001). At present, the radiative effects of aerosols have the largest uncertainties in global climate predictions to quantify climate forcing due to man-made changes in the composition of the atmosphere (IPCC, 1995). A better understanding of the formation, composition and

transformation of aerosols in the atmosphere is of critical importance in order to better quantify these effects.

Aerosols in the atmosphere have direct and indirect effects on the earth's climate. The direct effect is due to their optical and chemical properties. The cooling effect that aerosols have on the surface of the earth due to direct reflection of solar radiation is referred to as the direct climate forcing. Aerosols act to scatter or absorb solar and terrestrial solar radiation. The level of scattering depends on their physical and chemical characteristics. Consequently, aerosols modify the earth's incoming and emitted radiation and thus influence the warming and cooling of the planet. Aerosols can also act to modify the properties of clouds.

Aerosols also influence the size, abundance, and the rate of production of cloud droplets. Thus they influence cloud cover, cloud albedo and cloud lifetime. The effects of aerosols on the radiative properties of the earth's cloud cover are referred to as indirect effects, or indirect climate forcing. The liquid cloud forcing consist of two parts, the first indirect effect (change in droplet number associated with increases in aerosols) and the second indirect effect (change in precipitation efficiency associated with increases in aerosols). Some of the aerosols in the ambient air act as cloud condensation nuclei (CCN). The formation of cloud droplets is promoted by the presence of aerosol particles, and as a result, cloud formation occurs at lower super-saturation levels than they would if there were no aerosols (Houghton et al., 1994).

In addition to climatic effects, fine particulate aerosols are also linked with various health problems; from a runny nose and coughing, to bronchitis, emphysema, asthma, and they can even lead to death (Bardana 1995). Ultra-fine aerosol particles may contain excessive amounts of harmful substances that can be easily inhaled and eventually enter into the human blood stream. These particles

can penetrate deeply into the lungs, and cause lung difficulties and permanent lung damage (Manahan, 1991).

People exposed to high concentrations of atmospheric aerosols for long periods increase their chances of having serious health problems such as cancer, damages to the immune system, neurological damage, reproductive problems such as reduced fertility and respiratory problems (Manahan, 1991).

1.2. RESEARCH PROBLEM AND OBJECTIVES

The aim of this study was to determine the number size distribution of particles with radii in the range of 0.01–0.84 μm , and to investigate the existence of new particle formation in the Savannah environment. The study was conducted in the North West Province at the Botsalano game reserve. The main reason for choosing Botsalano was to determine if there is particle formation and growth of aerosols in the place where there are no industrial sources of air pollution. The specific objectives were:

- a. To determine the relationship between particle formation and particle growth.
- b. To determine the relationship between particle diameter and concentration.
- c. To determine the relationship between meteorological parameters and the number size distribution of particles with radius in the range of 0.01–0.84 nm.

1.3. MEASUREMENT SITE

The experimental part of this study involved the determination of the number size distribution of submicron atmospheric aerosols in Mafikeng (Botsalano game reserve) using a differential mobility particle sizer. The project was undertaken in the Botsalano game reserve, which is in the central district of the North West

Province. The game reserve is situated at latitude 25.32'30.4"S and longitude 25.45'16"E. The Botsalano measurement station is surrounded by a small forest and there are no industrial or mining sites within a radius of about 15 km. Figure 1.1 shows the map of Botsalano.

In this figure which was taken from Laakso et al., 2008, with permission, the detailed map is an enlargement of the red square on the small map in the lower right hand corner. On the detailed map are shown the following: 1. Botsalano game reserve (measurement site); 2. Mafikeng, 3. Slurry (cement factory); 4. Ramatlabama (a small village and chicken farm); 5. Zeerust; 6. Khunotswane; 7. Dinokana; 8. Lobatse municipal waste burning area. Directions and distances from the measurement site to some major regional pollution sources are shown. More details around the measurement site are shown in figure 1.2 also taken from Laakso et al., 2008, with permission.

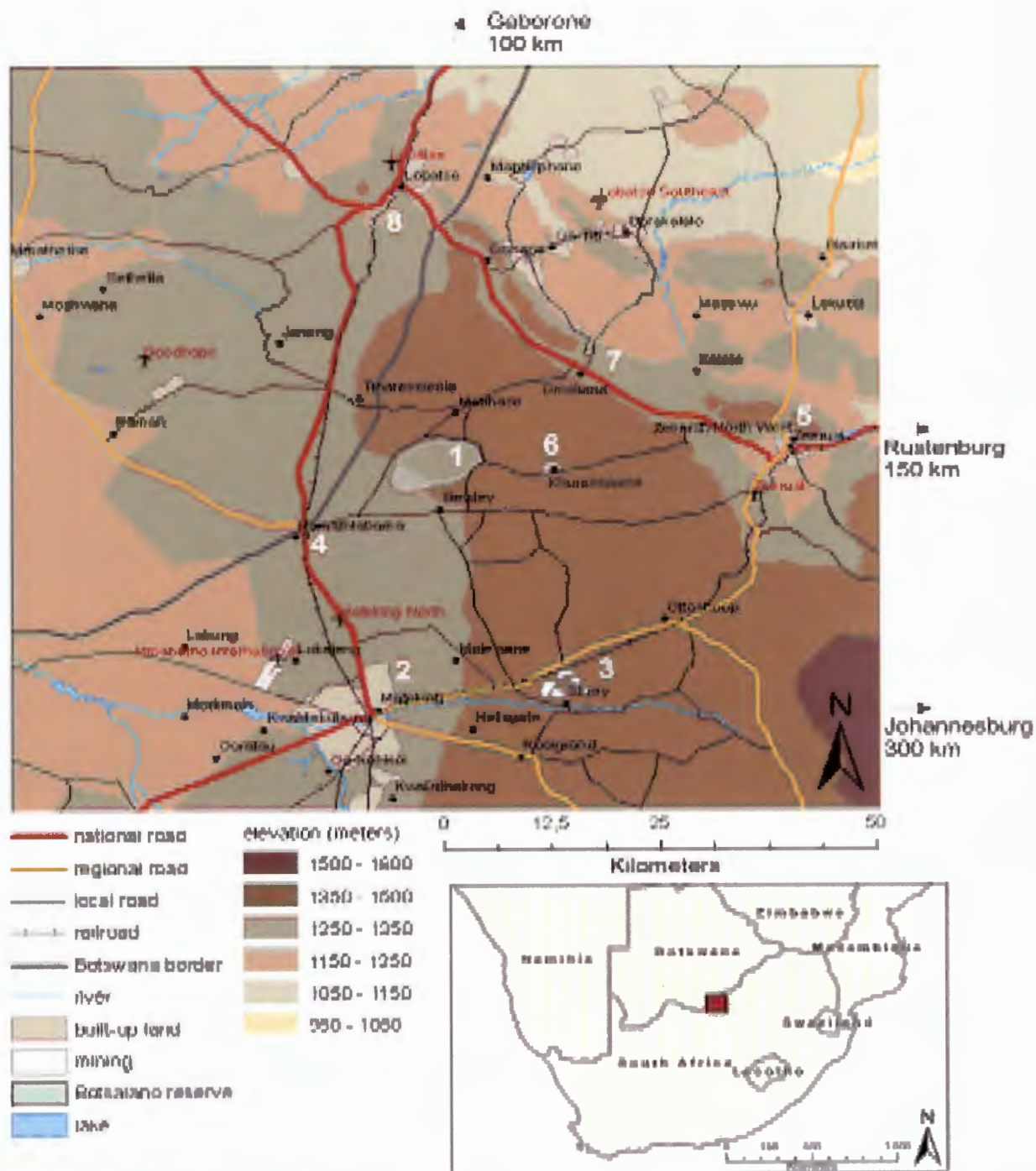


Figure 1.1: Map of Botswana

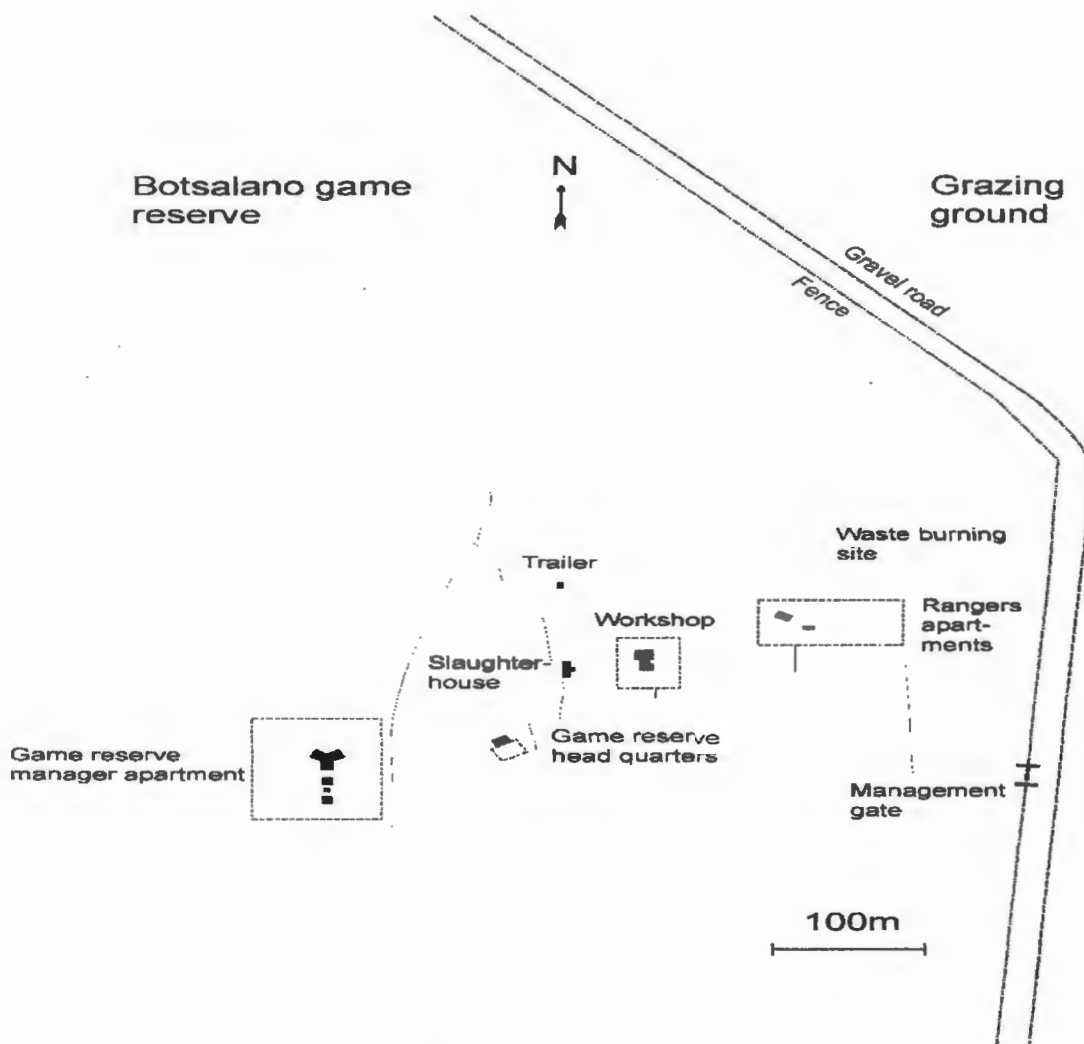


Figure1.2:Measurement site. All roads on the map are gravel roads (Map takenfrom Laakso et al., 2008, with permission)

CHAPTER 2

LITERATURE REVIEW

2.1. SOURCES OF AEROSOL PARTICLES

Atmospheric aerosols come from several local and regional sources. Local sources include vehicular emissions, wood burning and industrial processes. These sources can lead to urban air pollution (Fenger et al., 2002) and possible adverse health effects (Dockery et al., 1993; Harrison and Yin, 2000). Regionally, aerosols can be transported from areas of high emissions to relatively clean remote regions.

The natural sources include windborne dust, sea spray, volcanic activities and biomass burning, while emissions of particles attributable to anthropogenic activities arise from four sources namely: fuel combustion, industrial processes, non-industrial fugitive sources (e.g. construction work), and transportation sources (e.g. automobiles). Natural aerosols are probably 4 to 5 times larger than anthropogenic ones on a global scale, but regional variations in man-made pollution may change this ratio significantly in certain areas, like industrialized areas (Seinfeld and Pandis, 1998).

Aerosols can be directly emitted as particles (primary aerosols) into the atmosphere by volcanoes; in addition they can be generated through the effect of wind lifting dust particles in arid regions, from combustion during biomass burning, from sea spray and from vegetation. They can also be the result of chemical reactions (gas-to-particle conversion), and these are called secondary aerosols.

2.2. PROPERTIES OF AEROSOLS

Aerosols in the atmosphere have direct and indirect effects on the Earth's climate. The effect is related to their interactions with the radiation in the atmosphere, and these interactions are controlled by such properties as particle size, shape, chemical composition, amount and the time they survive in the atmosphere. One of the important parameters used to characterize aerosols is the size distribution which describes the number of particles observed to have a certain radius, for various size ranges.

2.2.1. PARTICLE SIZE AND CLASSIFICATION

Depending on their size, aerosols are broadly classified into three categories namely the Nucleation mode (radius 0.001 to 0.1 μm), Accumulation mode (radius ~ 0.1 to 1 μm) and Coarse particle mode (radius $> 1 \mu\text{m}$) (Hinds, 1999). Aerosols of different size ranges are caused by different atmospheric processes (Seinfeld and Pandis, 1998). The nucleation mode aerosols are the result of a nucleation process known as gas-to-particles conversion in the atmosphere. The accumulation mode aerosols are produced by coagulation (a process in which two or more aerosol are combined together to form a single aerosol), coarse mode aerosols are produced directly by mechanical processes (e.g. action of wind on aerosols). The accumulation mode aerosols significantly influence the visible solar radiation while the coarse mode aerosols can act as cloud condensation nuclei which lead to the formation of clouds (Parker 1999).

Particles in the atmosphere have widely differing shapes for which geometrical diameters are often not meaningful. Some means of expressing the size of such particles is essential since many important properties of the particles such as volume, mass, and velocity depend on size (Hinds, 1999). The size of irregularly shaped particles is expressed in terms of an equivalent diameter that depends on

a physical, rather than a geometrical property. An equivalent diameter is defined as the diameter of a sphere that would have the same value of a particular physical property as that of the irregular particle (Hinds, 1999).

There are different types of equivalent diameters. One of the most commonly used is the aerodynamic diameter, D_a , which is defined as the diameter of a sphere of unit density (1 g cm^{-3}) that has the same settling velocity in air as the particle under consideration. Aerodynamic diameter standardizes particles of various shapes and densities to spheres having the same aerodynamic property, namely the settling velocity. The particle aerodynamic diameter D_a is given by equation 2.1:-

$$D_a = D_g K \sqrt{\frac{\rho_p}{\rho_0}} \quad (2.1)$$

where D_g is the particle geometric diameter, ρ_p is the density of the particle, ρ_0 is the reference density (1 g cm^{-3}), and k is a shape factor, which is 1.0 in the case of a sphere. Because of the effect of particle density on the aerodynamic diameter, a spherical particle of high density will have a larger aerodynamic diameter than its geometric diameter. However, the density for most substances is less than 10 so that the difference between the geometric and aerodynamic diameters of a spherical particle is less than a factor of ~ 3 .

An important parameter in describing aerosol properties is the particle size. The diameters of atmospheric aerosols span over four orders of magnitude, from a few nanometers to around $100 \text{ }\mu\text{m}$. Atmospheric aerosols are normally classified into separate modes according to their size, formation process and atmospheric age. Based on the number, surface area, and volume distributions of aerosol particles shown in Figure 2.1, four distinct groups of atmospheric particles can be defined. Particles with diameters larger than $1.0 \text{ }\mu\text{m}$ are identified as the coarse

mode. These particles are mainly produced by mechanical processes and are introduced directly into the atmosphere from both natural and anthropogenic sources. The most significant source is the bursting of bubbles in the ocean, which creates coarse particles of sea salt. The wind also picks up dust, soil and biological particles and suspends them in the atmosphere (Hinds, 1999).

Particles with diameters between 0.1-1.0 μm are considered as the accumulation mode representing a region of particle growth mainly due to the coagulation of particles with diameters smaller than 0.1 μm and from condensation of vapours onto existing particles, causing them to grow into this size range. They can also be introduced directly into the atmosphere, mainly through the incomplete combustion of wood, oil, coal, gasoline and other fuels. Because of the nature of their sources, particles in the accumulation size range generally contain substantial amounts of organic material as well as soluble inorganics such as ammonium nitrates and ammonium sulphates. The accumulation mode is so named because particle removal mechanisms are least efficient in this regime, causing particles to accumulate there until they are ultimately lost through rain or other forms of precipitation (wet deposition) (Lindberg and Lovett, 1985).

The Aitken mode particles, extending from 0.01 to 0.1 μm in diameter, are formed from ambient-temperature gas-to-particle conversion as well as condensation of hot vapours during combustion processes. These particles act as nuclei for the condensation of low vapour pressure gaseous species, causing them to grow into the accumulation range. The lifetime of these particles is short, as they are lost principally by coagulation with larger particles. The Aitken and accumulation mode particles are referred to collectively as fine particles.

The final type of particles are those with diameters smaller than 0.01 μm shown in Figure 2.1 and are known as ultrafine particles. They are thought to be generated by gas-to-particle conversion processes that are not yet well understood at the molecular level. Because of their very small size and mass, they are difficult to

study given the available measurement techniques. These particles have been observed in bursts of very large numbers in the presence of either biogenic or anthropogenic emissions under favorable local conditions in many different environments including forests (Kulmala et al., 2004, 2004a) and coastal zones (O'Dowd et al., 2002) and have been referred to as the nucleation mode. These particles are only observed as a distinct mode at their source and have a very short lifetime, sometimes of the order of minutes, due to their rapid coagulation or random impaction onto surfaces.

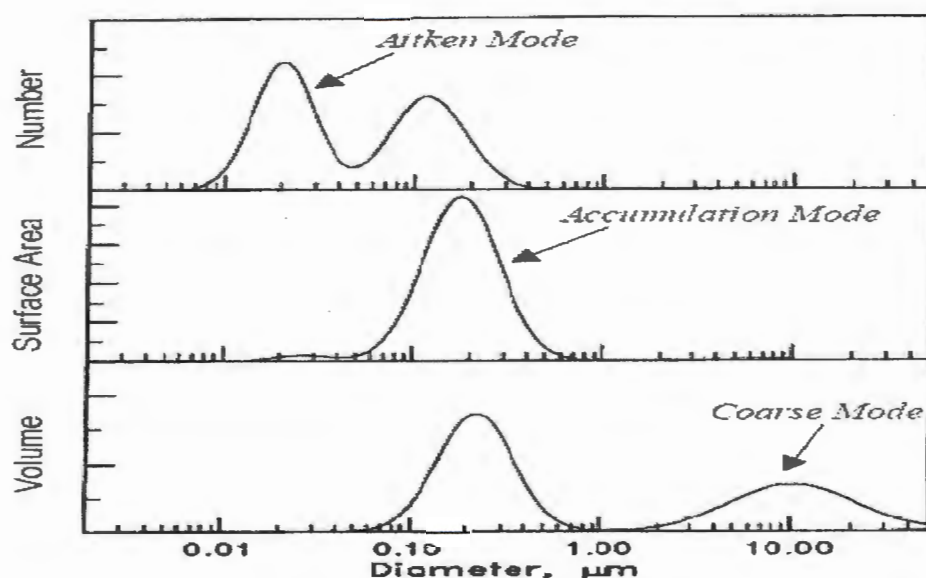


Figure 2.1: Number, surface area, and volume distributions of aerosol (Seinfeld and Pandis, 1998)

The nucleation and Aitken mode particles account for the majority of atmospheric particles by number, but due to their small sizes, they rarely account for more than a few percent of the total mass. Hence if the toxicological effects are determined primarily by the number of particles, rather than their mass, these small particles could ultimately prove to be of high importance. Accumulation mode particles generally account for a significant fraction of the total aerosol mass and have the greatest surface area. This makes these particles to be of significant importance to gas phase deposition and atmospheric heterogeneous chemistry. Most of the aerosol mass is found in the coarse mode, where large

particles contribute significantly to the optical properties of atmospheric aerosols. These modes, in general, originate and are transformed separately, and aerosols are removed from the atmosphere by different mechanisms. They have different lifetimes, chemical composition, optical properties and differ significantly in their deposition patterns in the respiratory tract. Therefore, the distinction between nucleation, Aitken, accumulation and coarse particles is a fundamental one in any discussion of the physics, chemistry, measurement, or health effects of aerosols.

2.2.2. CHEMICAL COMPOSITION

The chemical composition of particles is multi-factorial and depends on particle sources as well as post-formation processes. The most important chemical properties of particles include elemental composition, inorganic ions and carbonaceous compounds (organic and elemental carbon). Interest in the elemental composition, in general, derives from the potential health effects (Friedlander, 1970).

Particles arising from different formation mechanisms display significantly different properties both physical and chemical. In particular, there are substantial differences in the chemistry of primary and secondary particles. Primary particles generated from combustion processes are constituted mainly of soot, which is formed from hydrocarbons burning under fuel-rich conditions. Soot formed under such conditions appears most commonly as an ensemble of ultra-fine particles, but the size of the agglomerates can extend up to a few hundred nanometers. It is a gaseous-solid phase transition where the solid phase exhibits no unique chemical and physical structure (Bockhorn, 2000).

The natural biogenic hydrocarbons play an important role in the formation of tropospheric aerosols. The sunlight driven photo-oxidation of high-molecular

weight hydrocarbons has been shown to produce low vapour pressure reaction products that partition between the gaseous and aerosol phases. In the aerosol phase these reaction products are known as secondary organic aerosols. The natural biogenic hydrocarbons, might be effective sources (Hoffman et al, 1997), while the man-made hydrocarbons, aromatics are the most important sources (Odum et al, 1996).

2.3. PROCESSES AFFECTING AEROSOL PARTICLE POPULATION

Once airborne, particles can change their size and composition by condensation of vapour species, by evaporation, coagulation with other particles, by chemical reaction in the presence of water and super-saturation to become fog and cloud droplets.

Particles are eventually removed from the atmosphere by two mechanisms namely deposition at the earth's surface (dry deposition) and incorporation into cloud droplets during the formation of precipitation (wet deposition). Wet and dry deposition, lead to relatively short residence times in the troposphere, and because the geographical distribution of particle sources is highly non-uniform, tropospheric aerosols vary widely in concentration and composition over the earth.



Most of the aerosol sources are located near the earth's surface and hence their concentration (mass per unit volume) is larger near the surface, whereas atmospheric trace gases have lifetimes ranging from less than a second to a century or more. Residence times of particles in the troposphere vary only from a few days to a few weeks; they cannot remain suspended in the air for a long time because of gravity (Seinfeld and Pandis, 1998).

2.3.1. NUCLEATION

Nucleation plays a fundamental role whenever condensation, precipitation, crystallization, sublimation, boiling or freezing occurs. It is the starting point of the first order phase transition. Once the initial nucleation step has occurred, the nuclei tend to grow rather rapidly. Nucleation mode is formed by nucleation of atmospheric gases in a supersaturated atmosphere and their sizes are of the order of nanometers. The nucleation can occur from liquid phase to a solid phase (crystallization) or from liquid phase to a vapour phase (bubble formation). It occurs in the absence or presence of foreign materials. There are two types of nucleation processes, and these are homogeneous nucleation and heterogeneous nucleation. Homogeneous nucleation is a nucleation of vapour on embryos comprised of vapour molecules only in the absence of foreign substances. Heterogeneous nucleation is the nucleation on a foreign substance or surface such as an ion or solid particle. In addition, nucleation processes can be homo-molecular (involving a single species) or hetero-molecular (involving two or more species) (Parker 1999).

Homogeneous nucleation is a process in which one or more vapours form a cluster without a seed particle or ion (Springer, 1978). A stable thermodynamic system is formed when the number of molecules exceeds a critical value (critical radius). A critical number of the molecules depend on the concentration of the nucleating vapours and their temperature and pressure, while homogeneous nucleation may potentially increase both the number and the mass of aerosol particles per unit volume in the atmosphere. Heterogeneous nucleation can only increase the mass (Seinfeld and Pandis, 1998).

Heterogeneous nucleation is a process in which vapours nucleate on the surface of a pre-existing particle. Energetically, heterogeneous nucleation is often but not always more favorable than homogeneous nucleation (Fletcher, 1999). However, heterogeneous nucleation only increases the particle size and mass but not the

number concentrations in contrast with homogeneous, ion-induced or kinetic nucleation. The difference between heterogeneous nucleation and condensation is that, in the case of heterogeneous nucleation, there is a barrier preventing condensation. When this barrier has been exceeded, the phenomenon is called condensation (Seinfeld and Pandis, 1998).

2.3.2. CONDENSATION

Condensation is a process where vapour is deposited on a particle as it undergoes a phase transition from gas to liquid phase. This process usually requires a supersaturated vapour. It is initiated by the presence of small particles (nuclei) or ions that serve as condensation sites for particle formation (Kerminen et al., 2004). The super saturation for formation of atmospheric clouds is produced by cooling a saturated vapor by adiabatic expansion. It is a crucial factor governing particle formation and growth. Whatever the mechanism via which the particles are formed, they need to grow, and if the growth is too slow, the clusters are scavenged by larger particles (Seinfeld and Pandis, 1998). The reverse of growth by condensation is the closely related process of evaporation.

2.3.3. COAGULATION

Coagulation is a process where two separate particles collide and form a new single particle (Seinfeld and Pandis, 1998). The coagulation rate of particle population depends on the particle concentration. The net result is a continuous decrease in the number concentration coupled with an increase in particle size. Coagulation is the most important inter-particle phenomenon for aerosols. The coagulation efficiency in turn depends on the relative velocity of the colliding particles and their interceptions cross-sectional area, size, carrier gas, temperature and pressure. The nanometer-sized particles are most susceptible to coagulation scavenging, and Brownian coagulation is one of the key processes determining whether particles produced by nucleation ever reach

detectable sizes. The material of the particles may also affect the sticking probability of the particles, i.e. the probability that two colliding particles really form a stable particle together.

Aerosol particles suspended in fluid may come into contact because of their Brownian motion or as a result of their motion produced by hydrodynamic, electrical, gravitational, or other forces. The Brownian coagulation is often referred to as thermal coagulation (Springer, 1978).

2.3.4. DRY AND WET DEPOSITION

Deposition is a particle removal mechanism which reduces the particulate mass as well as the number in the surface. Dry deposition transports aerosol particles into surfaces without precipitation and removes most efficiently particles smaller than 50 nm and larger than a few micrometers in size; the former by means of Brownian diffusion and the latter by means of inertial impaction or gravitational settling. In addition to particle size, the particle removal efficiency is dependent on the nature of the underlying surface and is more effective over rough surfaces, such as forests and urban areas, than over smooth ones, such as water and grass lands (Rissler et al., 2004). While dry deposition acts on the particles continuously, wet deposition shows distinct peaks of rapid particle removal as a result of rain, snow or cloud droplet scavenging, and following deposition to the surfaces (Lindberg and Lovett, 1985).

Nucleation scavenging inside the cloud affects only particles large enough to activate during cloud formation although smaller inactivated particles may be incorporated in cloud water if they collide with cloud droplets. Below the clouds, the removal efficiency of particles by rain drops shows a minimum at the accumulation mode size range similar to dry deposition (Huff, 1975). A detailed quantification of the deposition mechanisms for modeling purposes is often

challenging because of the complex atmospheric interactions affecting the phenomena. Although size-resolved dry deposition models exist for different kinds of surfaces, it is difficult to account for the effect of the underlying surface properties or the small scale atmospheric motion in detail (Zhang et al, 2004).

The knowledge of many of the distinct mechanisms contributing to particle size distribution through wet deposition is not sufficient to allow their detailed description in numerical models. Although many models choose to simulate in-cloud and below-cloud deposition separate from each other, it has been found that a simple scheme not differentiating between these two and assuming a constant scavenging efficiency throughout the precipitation column, reproduced the observed fluxes of ^{210}Pb particle annual deposition as well as the more detailed schemes compared (Rannik et al., 2003).

2.4.HEALTH EFFECTS OF ATMOSPHERIC AEROSOLS

In the past, air quality standards for particulate matter were expressed in terms of mass of total suspended particulate matter (TSP).The standard (TSP)was then changed to mass suspended particulate matter less than $10\mu\text{m}$ in size also known as PM_{10} , and more recently it was modified to include particulate matter less than $2.5\mu\text{m}$ diameter $\text{PM}_{2.5}$ (EPA,1997). The rationale for basing air quality standards on smaller particles was linked to the fact that they can penetrate deeper into the lungs and because of this, they have a greater potential for harmful effects. In addition, they have larger surface-to-volume ratios and are commonly produced by gas-to-particle conversion or combustion processes, which are known to produce carcinogen aerosols such as aromatic hydrocarbons.

The large particles are removed in the mouth and throat, while the very small particles ($<0.10\text{ mm}$) are taken deep into the lungs where they impact in the

many tiny air pockets of the soft lung tissue and some of them are transported directly into the blood stream. The particles that are not retained are small enough to navigate the air passages of the mouth and throat, and move slowly enough to be expelled when a person exhales. Unfortunately, the tiny particles that are retained deep in the lungs contain the highest concentrations of toxic substances (Klenz and Fedde, 1978). Atmospheric aerosols affect health by transporting poisonous substances into the respiratory tract.

Aerosol mass and number concentration are much greater in urban or industrialized regions. Epidemiological studies have shown that such elevated aerosol mass concentrations are associated with increased risks of mortality and morbidity. Elevated number concentrations of ultrafine aerosol particles < 100 nm in particular showed association with increased risks in mortality and morbidity in epidemiological studies.

Many epidemiological studies observed an association between particulate air pollution and human health (Dockery et al., 1993, Pope et al., 1995). An increase of 1.5% in the total daily mortality with an increase of $10 \mu\text{g.m}^{-3}$ in $\text{PM}_{2.5}$ concentration has been reported (Schwartz et al., 1996). Elevated particulate air pollution has been associated with declines in lung function and increases in respiratory symptoms (Dockery et al., 1993, Pope and Dockery 1992, Pope et al., 2002).

Fine particulates have been associated with lung cancer and cardiopulmonary mortality (Dockery et al., 1993, Pope et al., 1995, Schwartz et al., 1996, Pope et al., 2002). Each increase of $10 \mu\text{g.m}^{-3}$ elevation in fine particulate air pollution was associated with approximately a 6% to 8% increase in the risk of cardiopulmonary and lung cancer mortality, respectively.

Most of the health effect problems are primarily associated with high concentrations of sulphur oxide gases and dust particles (Noble and Prather,

1998). Photochemical air pollution has resulted in a high concentration of ozone and photochemical product and their associated impact on human health, which is a major issue worldwide (Finlayson-Pitts and Pitts, 1999) and (Pope et al., 2002). Short term inhalation of ultrafine particulate air pollution and ozone at concentration levels that occur in urban environments can cause acute conduct artery vasoconstriction in healthy adults (Brook et al., 2002).

The primary parameters that determine the health effects of aerosol particles are their concentration, size, structure, and chemical composition. These parameters however, vary from one place to another and in time.

CHAPTER 3

INSTRUMENTS

3.1 MEASUREMENT TRAILER

The equipment used was installed in a mobile measurement trailer Eurowagon 4500U. The dimensions of the trailer were length 4.5m, width 2.1m, height 2.3m and it weighed 2500kg. The trailer was donated by the University of Helsinki to the North West University Mafikeng campus. It was assembled in Hyytiälä, Finland and was equipped to measure the following parameters: -basic meteorological parameters, air ion size distribution from 0.4 to 40nm, aerosol particle number size distribution from 10 to 840nm, aerosol particle mass for three functions $<1\mu\text{m}$ (PM1), $<2.5\mu\text{m}$ (PM2.5) and $<10\mu\text{m}$ (PM10), trace gas concentration for ozone, sulphur dioxide, nitric oxide, nitrogen dioxide and carbon monoxide. Routine maintenance of the equipment was carried out every two weeks.



3.2 DIFFERENTIAL MOBILITY PARTICLE SIZER AND ITS COMPONENTS

In this study, the objectives set in section 1.2 were achieved by using a differential mobility particle sizer (DMPS). The DMPS is an instrument used to measure aerosol size spectra. It was used for measuring small particles of radius 0.01–0.84 μm , and was combined with a differential mobility analyzer (DMA) and condensation particle counter (CPC) to automatically count and size the aerosol particles. In the DMPS aerosols of ambient air were passed through the TSI-3010 CPC system which was coupled to a data logger and thence to a computer where sampling data was captured.

Applications of DMPS include atmospheric aerosol measurements, indoor air quality studies, laboratory aerosol research, characterization of emission

sources, particle characterization for semiconductor, micro-contamination studies, aerosol characterization for atomizers, nebulizers, and other aerosol generating devices used for aerosol drug delivery and medical applications.

The DMPS system used is illustrated in Figure 3.1 which shows the flow path of the particles from the atmosphere until they were counted in the CPC. The lower and upper cut-off diameters of the system were 8 nm and 500 nm, respectively, and the set up measured one full size distribution every 10 min thus giving 144 distributions a day. The inlet for the DMPS system was at a height of 2m above the ground, and was mounted on the wall of the measurement cabin. The DMPS itself was inside the trailer cabin at a temperature of 25°C, which at low atmospheric temperatures led to evaporation of water and possibly some other volatile compounds from the particles before they entered the instrument (Aalto et al., 2001; Jokinen and Mäkelä, 1997).

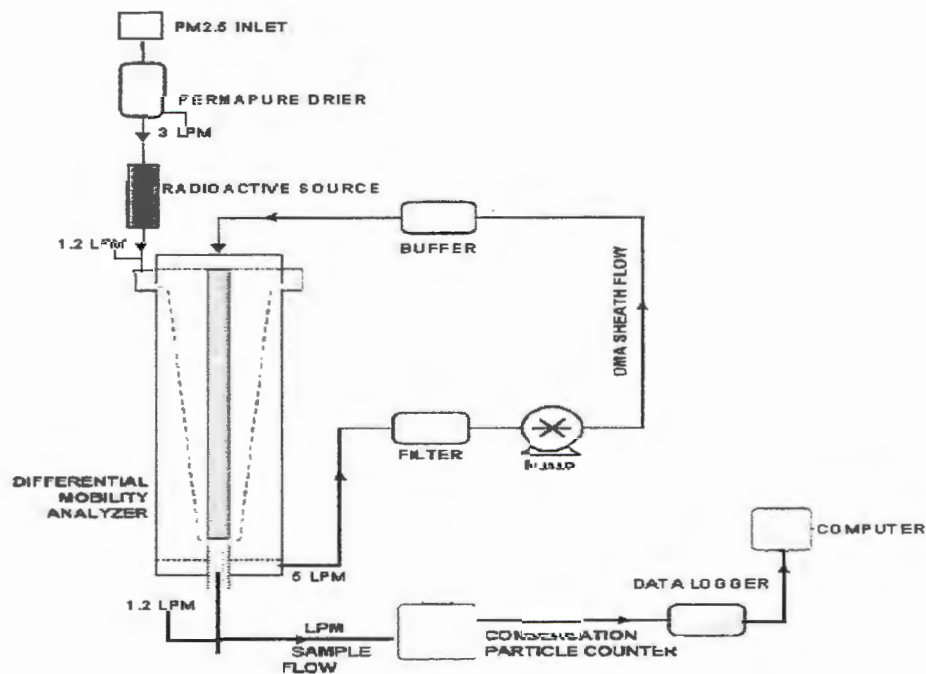


Figure 3.1: Structure of DMPS

3.2.1. DIFFERENTIAL MOBILITY ANALYZER

A Differential mobility analyzer (DMA) is an instrument that classifies charged particles according to their electrical mobility. The analysis is differential since only a small mobility fraction is classified at a time (Flagan, 1998). Basically the structure of the DMA instrument consists of two charged concentric cylinders with an inlet slit and a sampling slit. Aerosol particles for sizing are inserted into an annular region between the two cylinders at the inlet slit. They are carried by clean sheath air flowing through the annular region. Particles with mobilities in a certain narrow range are sampled at the sampling slit. Then an inversion calculation is done to infer the size distribution. The sizing depends on adjustable parameters such as the voltage between the cylinders and the flow rates. For sizing nanometer particles Brownian motion becomes important. The analysis of the trajectories of aerosol particles within the DMA is based on stream functions and depends on velocity and electric field distributions. A theoretical description of the instrument's performance has also been published (Knutson and Whitby, 1975).

The electrical mobility Z_p of a particle which is defined as the ratio of the constant limiting velocity a charged particle will reach in a uniform electric field to the magnitude of this field is described by the following equation, which is derived from Stokes' law:

$$Z_p = \frac{\text{Particle velocity}}{\text{Electric Field Strength}} = \frac{neC_c}{(3\pi\mu D_p)} \quad 3.1$$

Where n = number of charges on the particle, e = elementary charge, C_c = Cunningham slip correction, μ = dynamic viscosity of air and D_p = radius of particle.

3.2.2 CONDENSATION PARTICLE COUNTER

Condensation particle counters are instruments in which small particles are grown to optically detectable sizes by condensing some vapour on them. CPCs have been used in aerosol science for over a hundred years (Spurny, 2000). The CPC operates by drawing aerosol samples continuously through a heat saturator in which alcohol is vaporized and diffused into the sample stream. Aerosol sample and alcohol vapour pass into a cooled condenser where alcohol becomes supersaturated and ready to condense (McMurry, 2000). The most common type of CPC is the laminar flow CPC where the condensation expansion type growth is achieved by forced convection. The first models of CPCs of this kind were designed during the 70's by Rosen (Rosen et al., 1974), Bricard (Bricard et al., 1976) and Sinclair (Sinclair and Hoopes, 1975).

Particles present in the sample stream serve as condensation sites for alcohol vapor. Once condensation begins, the particles grow quickly into larger alcohol droplets and pass through an optical detector where they can be counted easily. Particle size range is 0.01-1.0 micrometer (10 to 1000 nanometers) and the concentration range is 0 to 100,000 particles per cubic centimeter (pt/cc) with about 20% accuracy (Jacobson and Turco, 1995). The temperature range used was 50° to 95° C. The units can be used either in survey mode or in logging mode. Logged data is downloaded to a computer using TSI's aerosol Instrument Manager Software. Readings are given in particles per cubic centimeter (pt/cc) and the unit is non-discriminatory for particular materials counting all particles passing through the detector (Bricard et al., 1976).

The system whose schematic diagram is shown in figure 3.2 includes a power supply which allows the analyzer to be powered from an ac supply. This enables the unit to be left in the logging mode for extended periods of time. The meter

uses high purity isopropyl alcohol and must remain horizontal (display facing up) during use or damage can occur to the unit. During transportation the alcohol must be removed from the unit to prevent leakage and damage to the optics inside the unit. Four green status lights on the right-hand side of the CPC front panel monitor the laser, the temperature, the flow and the liquid. An amber status light monitors the presence of particles (Rosen et al., 1974).

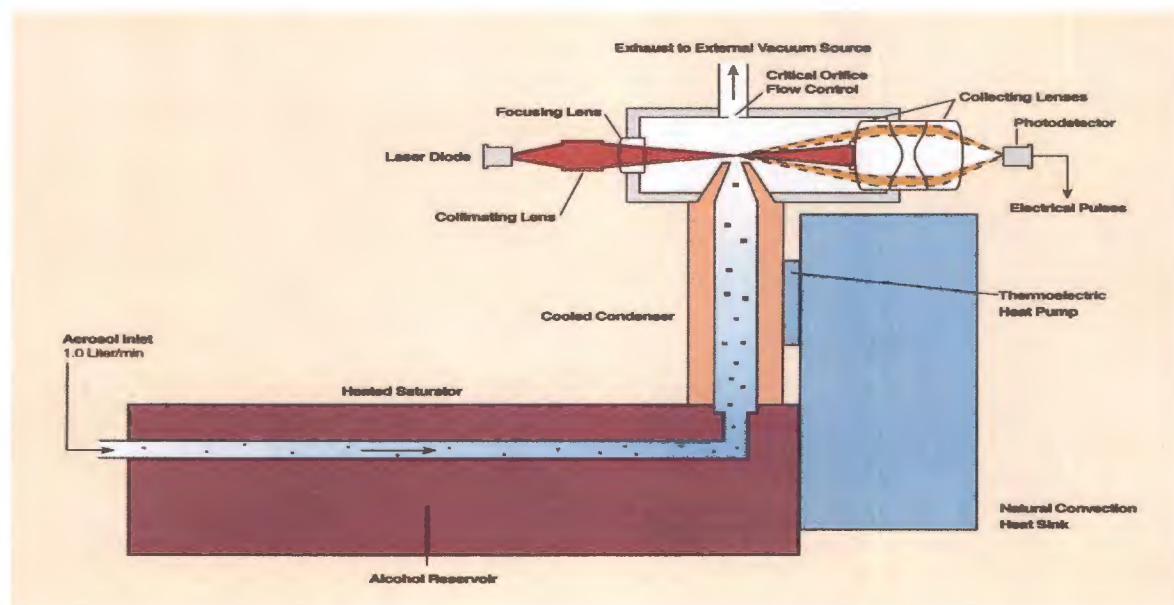


Figure 3.2: Schematic diagram of Condensation Particle Counter (Aalto et al., 2001).

When an air stream enters the condenser, it is cooled by a thermoelectric device outside the condenser. The vapour cools, becomes supersaturated, and begins to condense on the particle nuclei to form larger droplets. The droplets pass from the condenser tube, through a nozzle into the optical detector. The liquid that condenses on the walls of the condenser tube runs down into the saturator and is absorbed into the saturator and reused.

The counter sensor's focusing optics consists of a laser diode, a collimating lens, and a cylindrical lens. This combination forms a horizontal ribbon of laser light above the aerosol exit nozzle. The collecting optics incorporate a pair of aspheric

lenses that collect forward scattered light onto a low-noise photodiode. The main beam is blocked by a light-stop in the center of the first aspheric lens. Surface temperature of the optics housing is maintained by mechanical contact with a heat sink, at or above the saturator temperature to avoid condensation on the lens surfaces (McMurry, 2000).

3.3. PRINCIPLE OF OPERATION

The DMPS's operation is based on the principle that the mobility of a charged particle in an electric field depends on their size and electrical charge. Particles entering the system are neutralized (using a radioactive source). The neutralizer brings the particles into a well-known charge distribution. The result is a Boltzmann distribution of negatively and positively charged particles. The particles enter a DMA where the aerosols are classified according to their electrical mobility, with only particles in a narrow range of mobility exiting through the output slit. This mono-disperse distribution then goes to a CPC which determines the particle concentration at that size.

The DMA used comprised a cylinder with a negatively charged rod at the center. The main flow through the DMA is particle free 'sheath' air. It is important that this flow is laminar. The particle flow is injected at the outside edge of the DMA and particles with a positive charge move across the sheath flow towards the central rod at a rate determined by their electrical mobility. The size of particles exiting through the slit is determined by the particles size, charge, central rod voltage, and flow within the DMA (Winklmayr et al., 1991).

CHAPTER 4

EXPERIMENTAL PROCEDURE

4.1. MEASUREMENT PROCEDURE

The DMPS system was assembled as shown in Figure 3.1. The inlet to the DMPS which was on top of the trailer had a PM2.5 impactor attached to it to select from the incoming particles, only those with diameters $\leq 2.5 \text{ nm}$. Prior to taking data, the inlet was removed, opened and cleaned with propanol. The impactor attached to the inlet was greased with vacuum grease to ensure that particles with diameters greater than 2.5 nm were trapped inside it and did not proceed with the airflow to the DMA.

All instruments were connected together care being taken to ensure that there was no air leakage. Four flow rates were measured namely: the sheath flow, the drier flow, aerosol inflow and aerosol outflow from the DMA. The drier flow was 3 litres per minutes (LPM), sheath flow was 5 litres per minutes, aerosol inflow and outflow were 1.2 litres per minute each.

A zero test was done, the CPC power voltage was turned off or alternatively a filter was placed to the 3-way valve (CPC ample flow) and CPC was monitored to insure that it registered zero voltage after 5 minutes. If zero was not registered in the display screen of CPC, it meant that there was a fault somewhere. The power was switched on after a zero test was completed. The butanol bottle was checked to ensure that there was a sufficient amount, the DMPS used about 0.5 litres in two months. The equipment was turned on and the DMPS program was

started in the computer to record sampled data. To ensure the quality of data the maintenance of the equipment was made every two weeks.

4.2. DATA ANALYSIS

To analyze data, equation 4.1 was used to find the formation rate and particle growth. The number concentration based on size distribution can be calculated using the equation:

$$n_N(\log D_p) = \frac{dN}{d\log(D_p)} \quad (4.1)$$

where D_p represents the diameter of the particles, $n_N(\log D_p)$ is the particle number concentration and dN is the number of particles in an infinitesimal size range. From the DMPS plot of particle diameter as a function of time, two points were taken using selector (ginput (2)), and the maximum nucleation mode was labeled as d_p^2 . The particle formation rate J_p is given by equations 4.2 in which $N_{\max}$ and $N_{\min}$ are the maximum and minimum number of particles between 10nm and d_p^2 and were found from the DMPS plots.

$$J_p = \frac{\nabla N}{\nabla t} = \frac{(N_{\max} - N_{\min})}{(t_{\max} - t_{\min})} \quad (4.2)$$

where ($N_{\max}$ and $N_{\min}$) are the maximum and minimum number of particles between 10nm and d_p^2 respectively, and $t_{\max}$ and $t_{\min}$ are the maximum and minimum times during growth rate and particle formation. During the growth of particle from 10nm to d_p^2 some of the particles collide and form larger particles whose diameters can be between 10nm and 840 nm. This represents particle loss which has to be taken into account in equation 4.2. The loss can be approximated by calculating the mean size of (d_p^2 to 840nm) particles and (10nm to d_p^2) particles.

The particle formation rate J of aerosols in new particle formation event is given by:

$$J = \frac{dN}{dt} + F_{Coag} + F_{growth} \quad , \quad (4.3)$$

where N is number concentration of aerosol particle in the size range in analysis, F_{Coag} is flux of particle out from the size range due to coagulation to pre-existing particle and F_{growth} is the loss of particle due to growth out of the size range (Dal Maso et al., 2005). The dN/dt term is approximated for the nucleation event by fitting a line in the period when nucleation mode particle number concentration increases linearly. If nucleation mode size range is selected wide enough, the particles do not grow out of it during the period of new particle formation and then the F_{growth} can be neglected.

CHAPTER 5

RESULTS AND DISCUSSION

The results presented in this chapter are of measurements that were taken continuously and on a daily basis from July 2006 to July 2007 in Botsalano game reserve. It is worth noting that there are no studies conducted in Southern Africa or a similar environment that one could compare with, hence a few comparisons were made to studies elsewhere in the world.

5.1. OBSERVATION OF PARTICLE FORMATION AND GROWTH

The particle growth and formation can be observed from the banana curve in the plot of the relationship between particle diameter and the time of day. The particles losses can be observed where there are no banana curves.

5.1.1. DAILY VARIATIONS IN PARTICLE GROWTH AND FORMATION

The daily variations shown in Figures 5.1 were obtained from Monday 24th to Saturday 29th July 2006. The particle formation and growth are represented by the banana curves shown in Figure 5.1 (a). The results show that particle growth takes place mostly during the day. From the graph it is also observed that the formation of particles on the 26th and 27th of July was low compared to that on the 24th and 29th. Further, from the data of the graph displayed in Figure 5.1 (a) it was observed that particle losses occurred in the size range between 50 nm and 290 nm. The events of particle formation shown in Figure 5.1 (a) coincide with periods of high global radiation, high temperature and low relative humidity shown in the panels of Figure 5.1 (b). Similar effects of meteorological factors on

particle formation suggested by the graphs have also been observed in studies of seasonal variations of trace gases, meteorological parameters, and formation of aerosols in boreal forests (Yulia et al. 2005). Atmospheric fine particle formation events have been observed around the world in various environments from polluted cities to remote polar background areas (Kulmala et al., 2004).

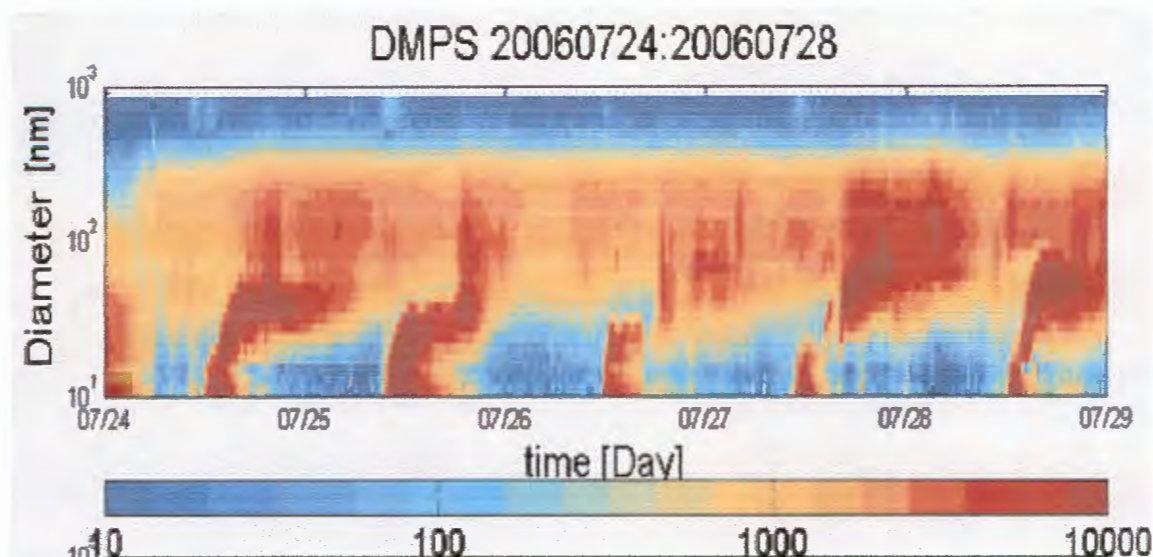


Figure 5.1: (a) Diameter of particle against time at growth

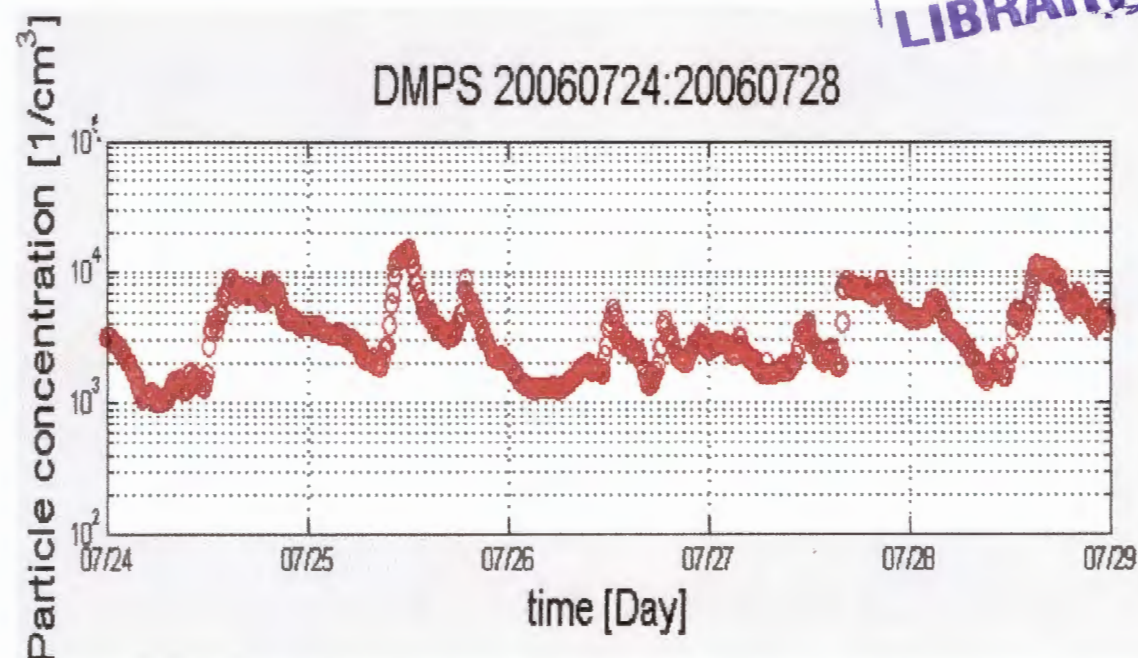


Figure 5.1:(b) Particle concentration against time at growth

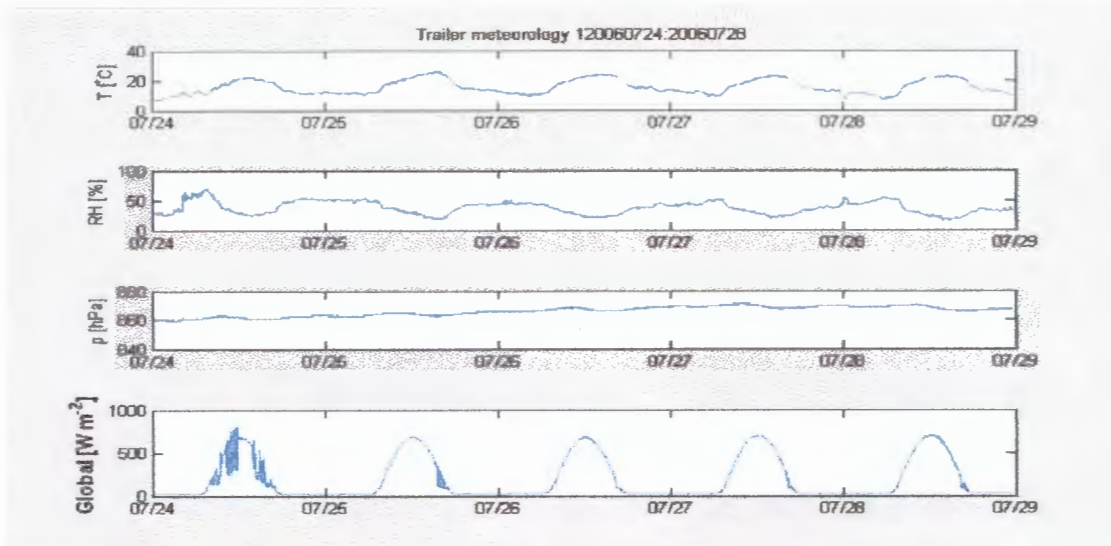


Figure 5.2: (a)Temperature, Relative Humidity, Pressure and Global Radiation from Monday 24th July to Friday 28th 2006.

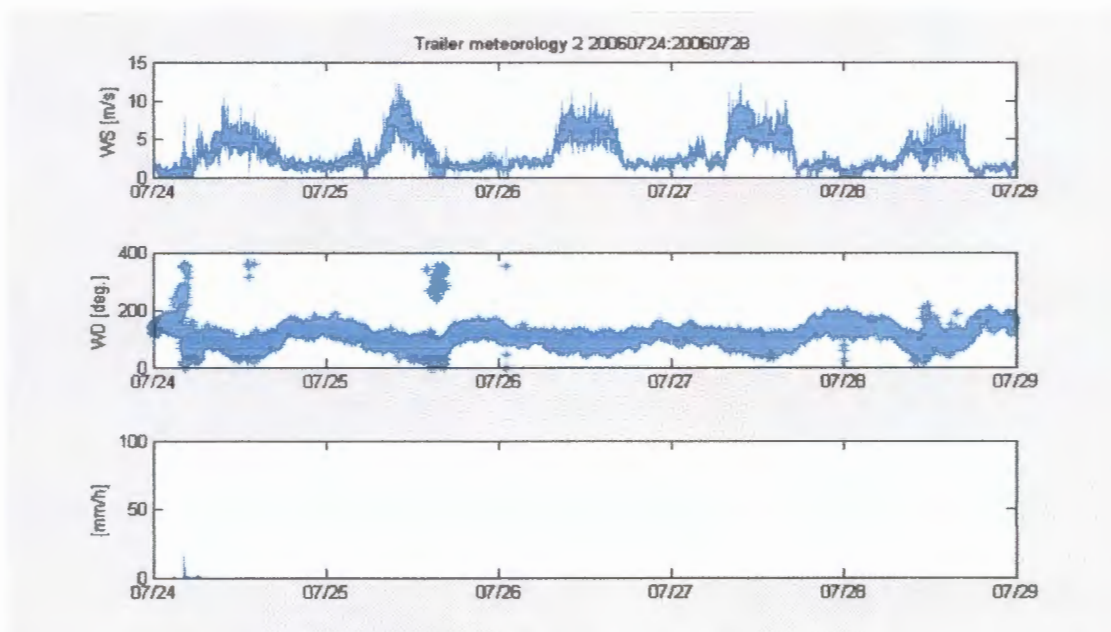


Figure 5.2: (b)Wind speed, wind direction and rainfall from Monday 24th July to Friday 28th 2006.

In Figure 5.2 (a), the relative humidity is shown to be around 60% during the night of the 24th, and it fluctuated around 50% from the 25th up until the 28th. The relative humidity was observed to decrease during the day when particle

formation increased. This can be explained by noting that some of the moisture content is used to enhance particle growth which leads to the low relative humidity recorded.

It has been observed (Lindberg and Lovett, 1985) that some particles are lost due to wet deposition which is a distinct peak of rapid particle removal resulting from rain and cloud droplets. The precipitation recorded in the bottom panel of Figure 5.2 (b) is proposed to be responsible for the low values of particle concentration in Figure 5.1 (b) around that time.

A comparison of Figure 5.1(a) and Figure 5.2 (b) shows that as wind speed increases, the particle formation and growth increases. The wind direction in the middle panel in Figure 5.2 (b) shows that the highest average wind was coming from the south-easterly direction, pollution from this direction may be due to traffic and some dust liberating process from the cement industry as can be seen from Figure 1.1. Particle formation depends on wind speed such that high wind speed yields high formation rate although it sometimes leads to dilution. It is observed from a comparison of Figure 5.1 (a) and the top panel of Figure 5.2 (a) that temperature has an impact on particle formation in that when the temperature is greater than 20°C, there will be particle formation and growth. The top and bottom panels of Figure 5.2 (a) also show that temperature and global radiation are positively correlated to particle formation in line with results from other studies (Yulia et al 2005).

5.1.2. HOURLY VARIATIONS OF PARTICLE GROWTH AND FORMATION

The hourly variations are useful in determining the specific hour in which particle formation was observed within a day. Figure 5.3(a) shows hourly variations in particle formation while Figure 5.3 (b) shows particle concentration for a typical winter day.

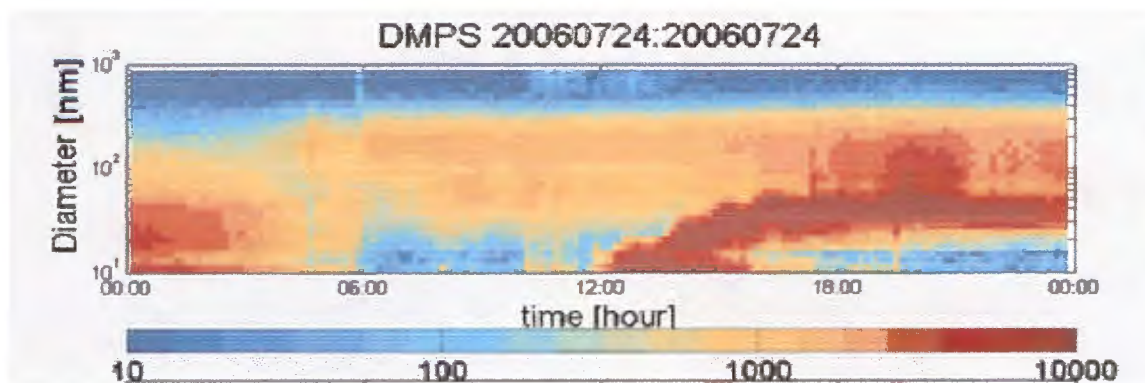


Figure 5.3: (a) Particle diameter against time for 24 July 2006

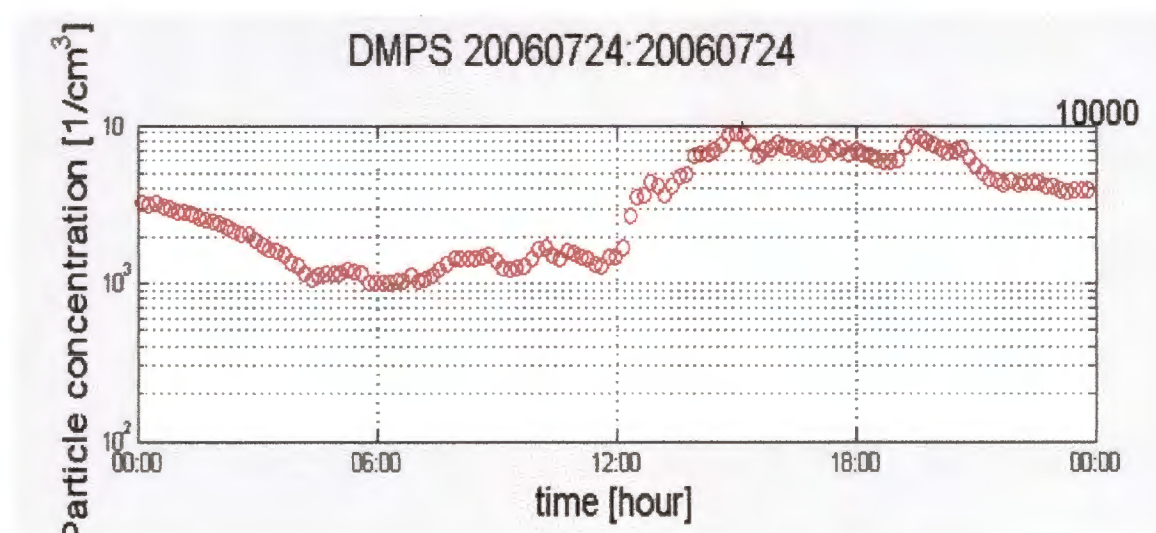


Figure 5.3:(b) Particle concentration against time for 24 July 2006.

Particle growth observed from Figure 5.3 (a) ranges between 10nm and 70nm and the particle concentration on Figure 5.3 (b) is in the range from 1000 to 10000 ($1/\text{cm}^3$). The particle formation and growth were observed around 12 hours, the same time that an increase in particle concentration was observed thus indicating a positive correlation between them.

Figures 5.4 (a) and (b) displaying data for a typical summer day show the same trend. The possible sources for the formation may be biogenic (related photosynthesis), sulphuric acid, or some other sulphur containing compounds.

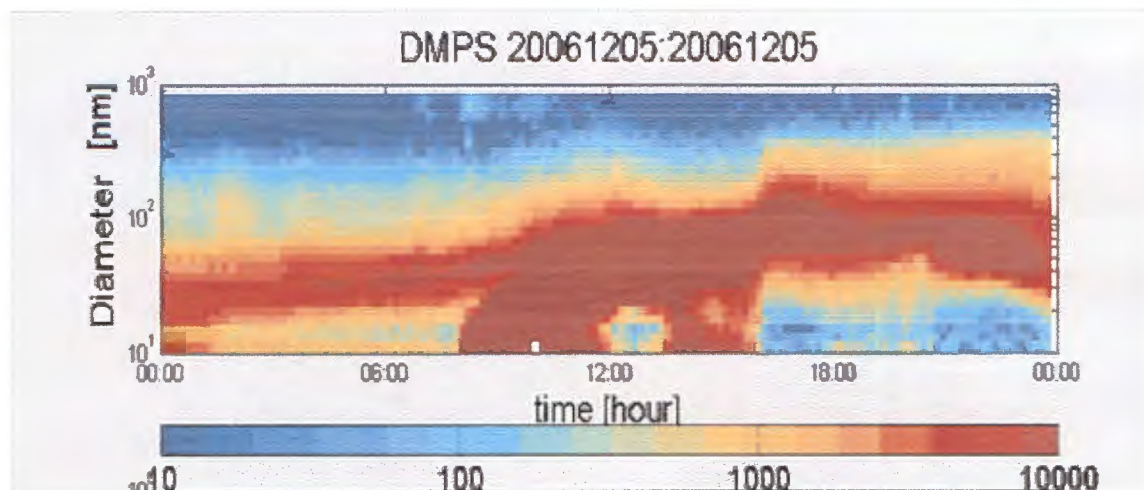


Figure 5.4: (a) Particle diameter against time for 5th December 2006.

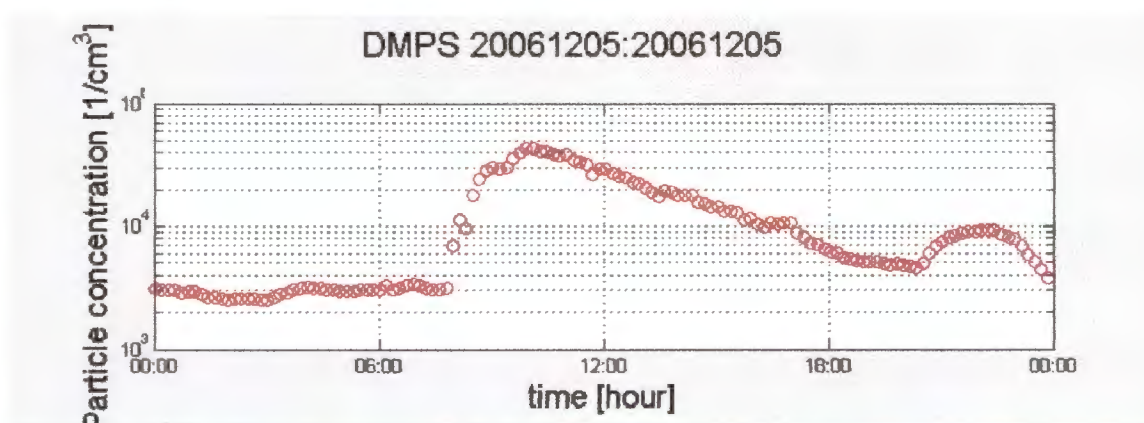


Figure 5.4:(b) Particle concentration against time for 5th December 2006.

The wind speed was very high around 13 m/s and this may imply that the particle growth is directly proportional to the wind speed. The wind direction data shows that most of the particles that contributed to the particle formation are from the south east of the measurement site; this could be due to traffic, biogenic process from the trees around there and dust particles from the cement industry.

Comparison of figure 5.3 (b) and Figure 5.4 (b) shows that in summer, the formation of particles started before 12h00 while in winter it started after 12h00 noon. These differences may be ascribed to differences in temperatures during the two seasons and the fact that in winter sunrise was later than in summer. In

a study of the comparison of new particle formation events at two locations in northern Finland, it was found that the difference in starting times for the events could be explained by the difference in the times the sun rose at the two locations (Komppula et al 2003).

The particle formation and growth events occur in the morning but at different times for different days (Kumala et al., 2004, Kulmala 2003). The day-time particle growth events are higher than those occurring in the afternoon and in the morning mainly due to temperature and global radiation differences.

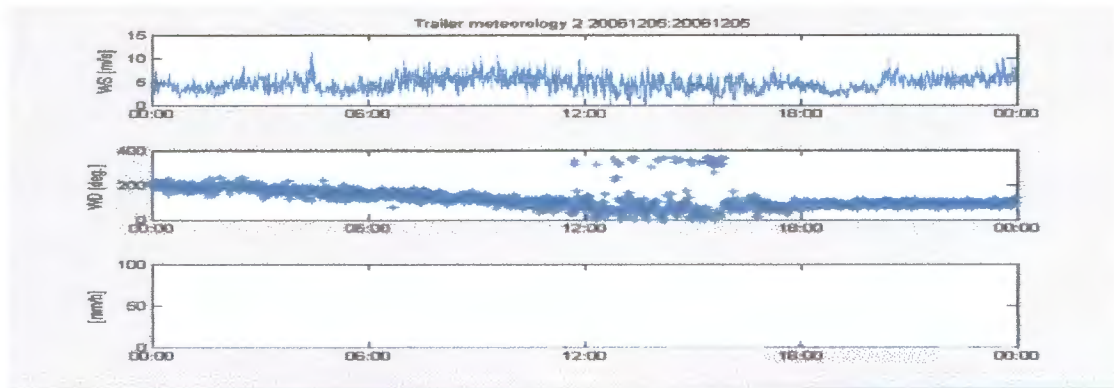


Figure 5.5 (a): The meteorological results for 5th of December 2006 for rain intensity, wind speed and wind direction.

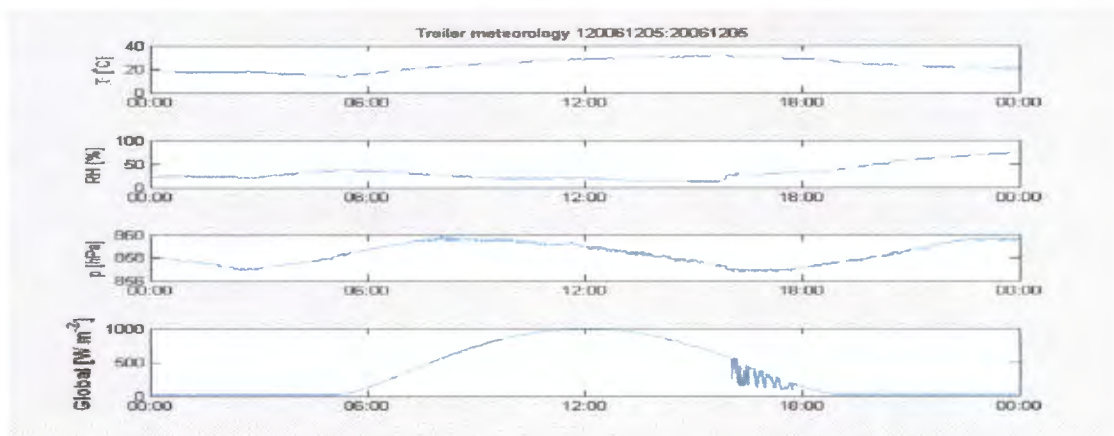


Figure 5.5 (b): The meteorological results for 5th of December 2006 for global radiation, relative humidity, pressure and temperature.

The rain normally reduces the concentration of particles. The lower concentration can be observed only when the relative humidity is over 98% (Hôrrak et al., 2003). The relative humidity in the data plotted in Figure 5.5 (a) was below 50% for most of the day, becoming lowest at the time when particle formation events took place. The particle concentration shown in Figure 5.4 (b) ranged from 3000 cm^{-3} to 70000 cm^{-3} . The high particle formation can be observed only when relative humidity is high (Laakso et al., 2004). The lower the particle concentration the lower the formation rate, and the higher the particle concentration rate the higher will be the formation rate.

5.1.3. WEEKDAY AND WEEKEND VARIATIONS OF PARTICLE GROWTH AND FORMATION

The weekdays and weekend variations in the particle growth give an indication of socio-economic activities from the nearest village and people working in the game reserve.

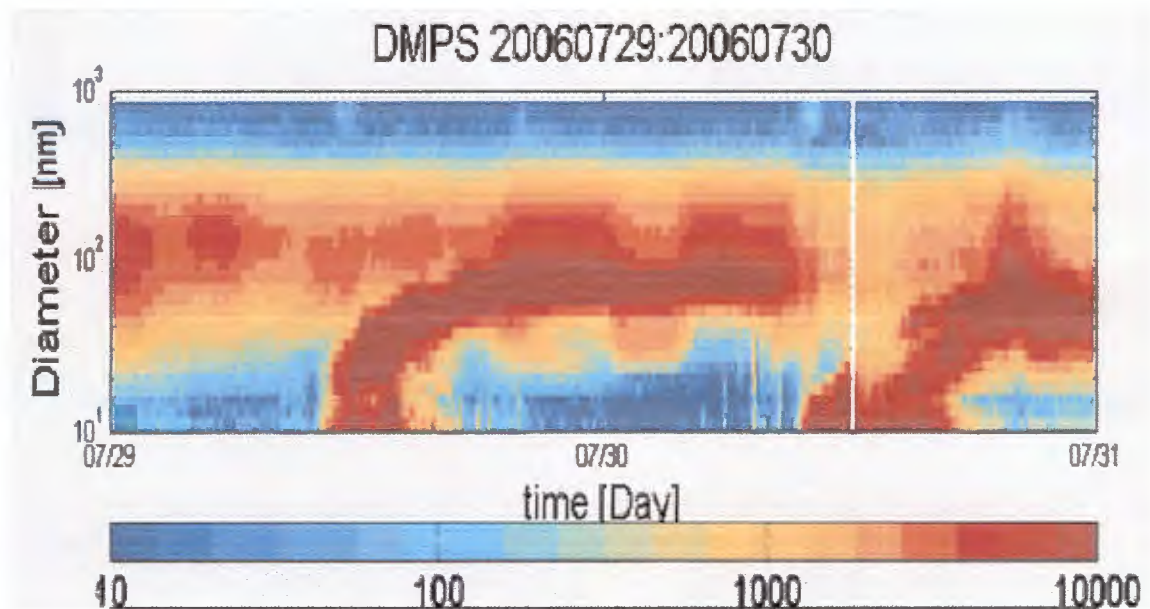


Figure 5.6: (a) The winter variation of particle growth during the weekend of 29th – 30th July 2006.

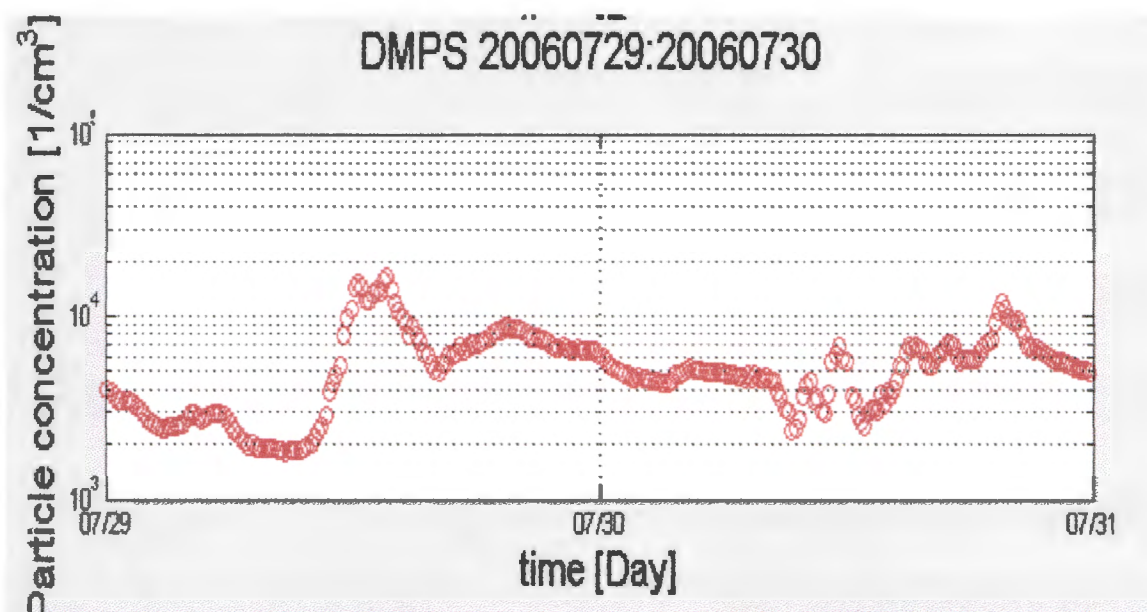


Figure 5.6:(b) Particle concentration (b) during the weekend of 29th – 30th July 2006.

The results shown in Figure 5.6(a) were obtained during the weekend of the 29th and the 30th of July 2006. The particle formation and concentration during the 30th was low when compared to the formation rate on the 29th. The formation rate on Sunday is the same as the formation rate observed during the week but on the Saturday the concentration from Figure 5.6(b) was very high as was also the formation rate of particle. This may be due to the traffic and the activities around the game reserve and biomass burning from the surrounding village.

The results obtained during this weekend show that on Saturday the 29th of July, particle concentration was higher than that obtained on the 30th of July. It was also observed that the values of global radiation, wind speed, temperature, pressure and relative humidity were different between these two days. These are shown in Figures 5.7 (a) and 5.7(b). Since all these meteorological parameters can affect particle formation rate and concentration they could be partly responsible for the different results of the two days.

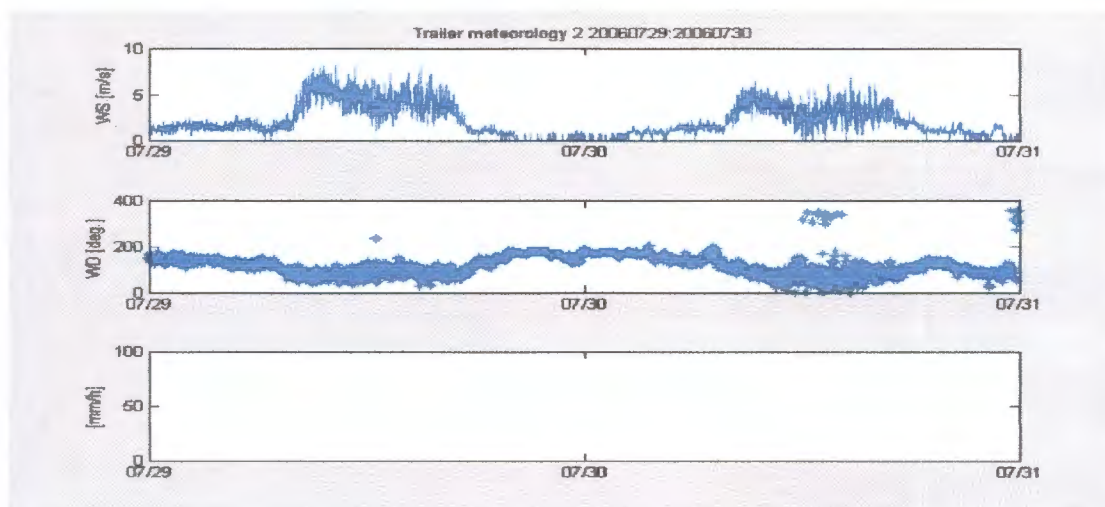


Figure 5.7 (a): Rain intensity, wind speed and direction on the 29th and the 30th July 2006.

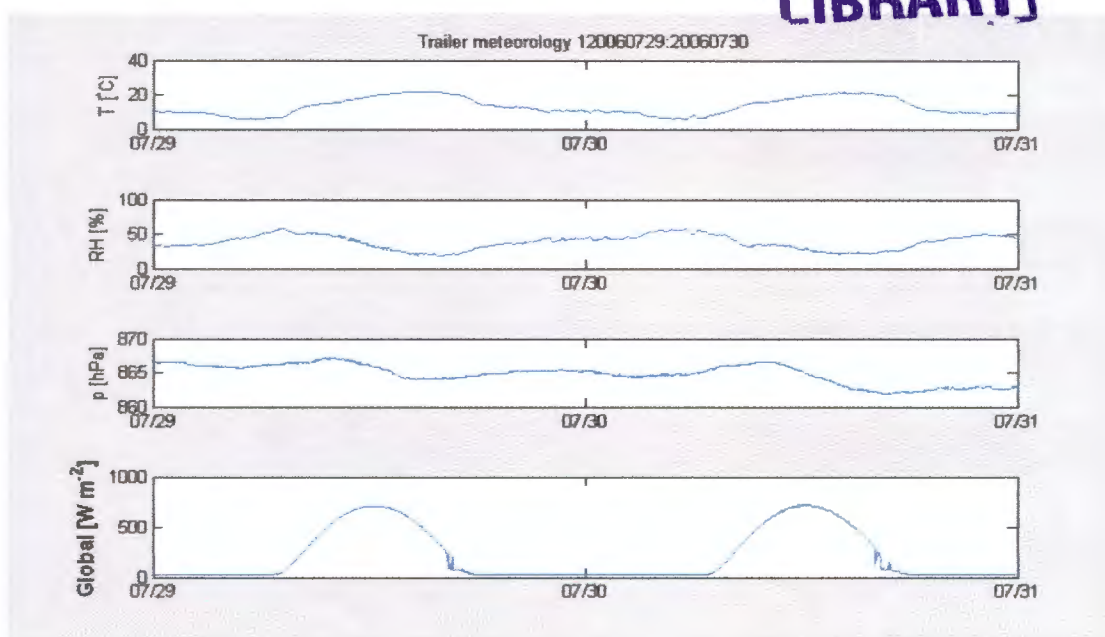


Figure 5.7 (b): Global radiation, relative humidity, pressure and temperature on the 29th and the 30th July 2006.

The average wind speed during the period when particle formation takes place was over 5 m/s and lower before and after the formation events. This tells us that particle formation increases when wind speed increases. The wind direction was below 90 ° during the particle formation events, and this tells us that the

pollution was coming from the eastern side of our measurement site. This may be due to traffic, organic compounds, biogenic materials coming out of Zeerust and Rustenburg which are large population areas to the east and north-east of the measurement site.

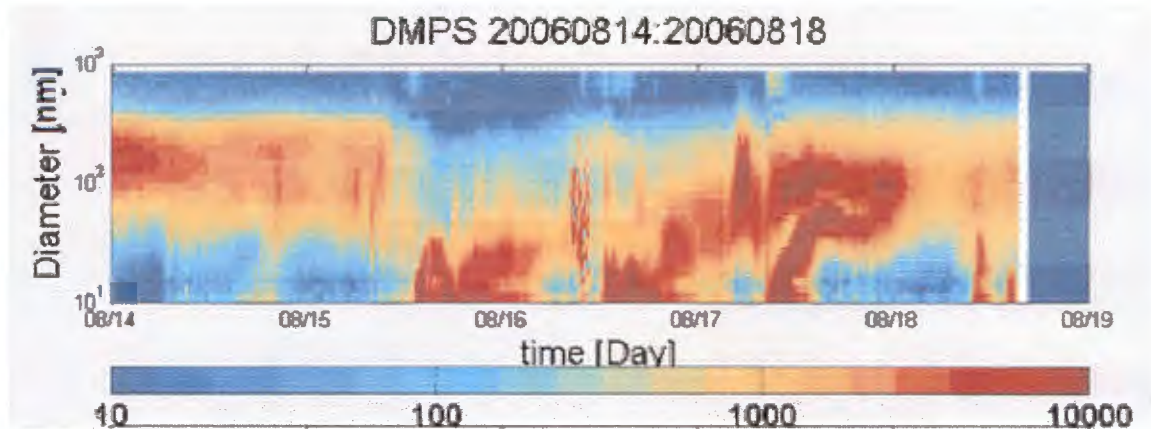


Figure 5.8: (a) Particle concentration for weekdays of 14-18 August 2006.

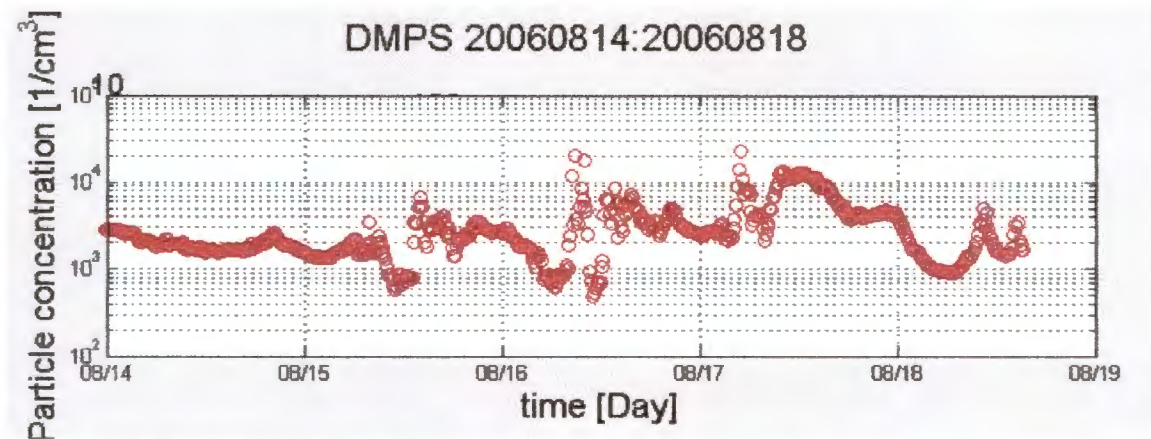


Figure 5.8:(b) Diameter of particles for weekdays of 14-18 August 2006

The weekday results for the week of 14th to 18th August 2006 are shown in Figure 5.8 (a) for particle formation and Figure 5.8 (b) for particle concentration. In August, the particle formation was very low on the 14th compared to the 15th, and as indicated in the graph, particle formation started at about 13h00. Particle concentrations on the 14th to 18th August were between 500 cm⁻³ and 30000 cm⁻³, and the growth rate was generally low. The diameter of particles on the

14th ranged between 10 nm and 20 nm. The diameter of particles on the 15th up until the 18th ranged from 10 to 40 nm. This may be due to particle growth related to concentrations of condensable vapours which were high resulting in making the particle concentration to be low.

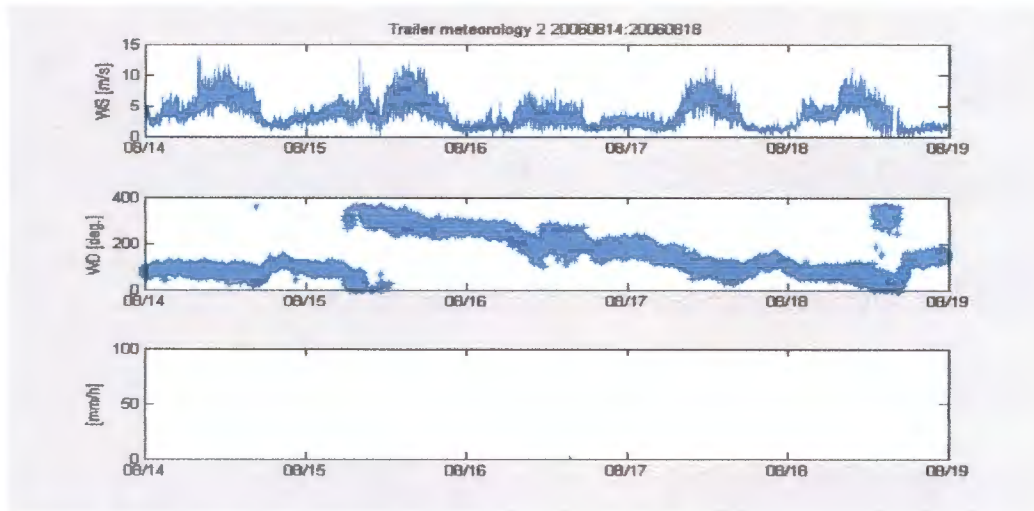


Figure 5.9 (a): The meteorological results from 14th to 18th of August 2006 for Rain intensity, wind speed and direction.

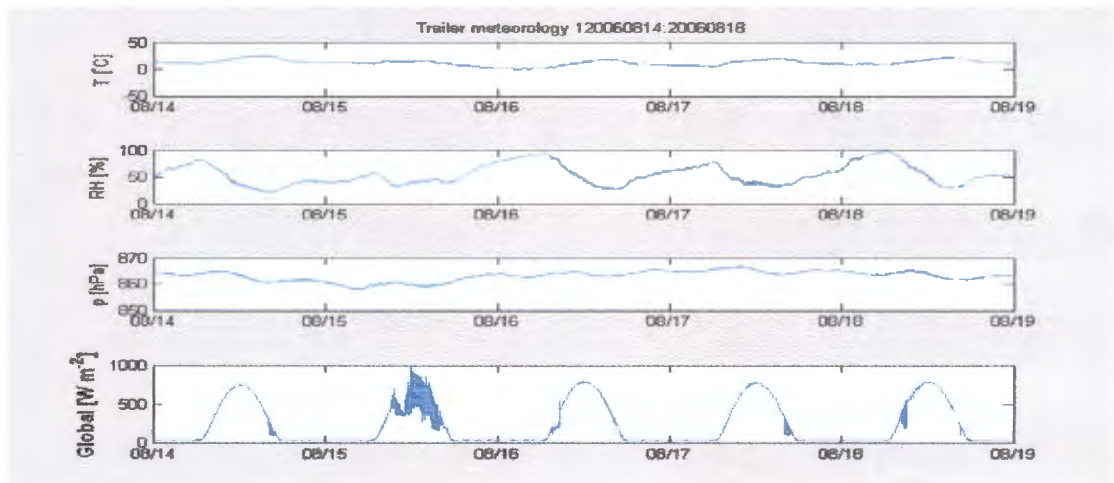


Figure 5.9 (b): The meteorological results from 14th to 18th of August 2006 for Global radiation, relative humidity, pressure and temperature.

5.1.4. SEASONAL VARIATIONS IN PARTICLE GROWTH AND FORMATION

The change in season affects the particle growth due to different meteorological conditions. The results presented in this section are those of data collected from July 2006 to February 2007 covering part of winter, spring and part of summer.

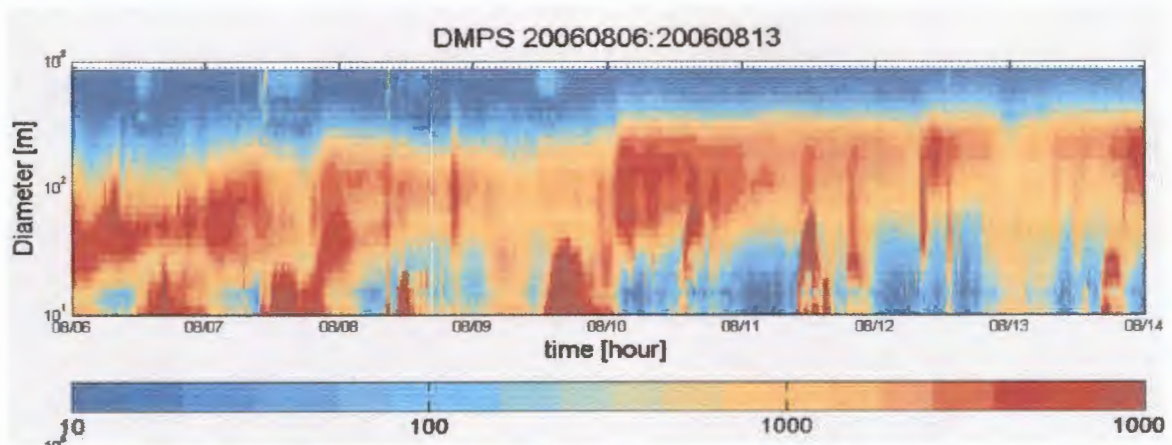


Figure 5.10: (a) Particle diameters against time for August 2006.

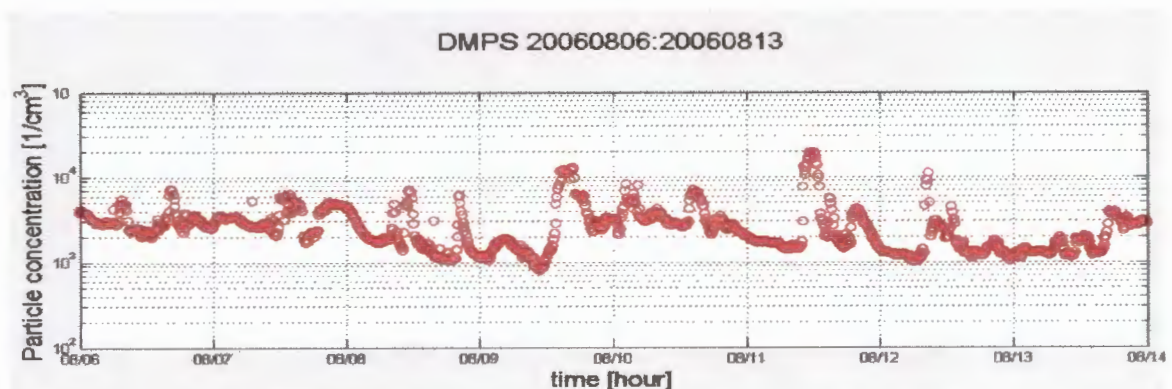


Figure 5.10:(b) Particle concentration against time for August 2006.

Figures 5.10 give results for one week in August which is part of winter. The data shows that in August 2006 particle formation was very weak (below 10000cm^{-3}) on the 9th and 11th when compared to the particle formation on the 10th, 12th and 13th (above 10000cm^{-3}). The particle diameters were in the range of 10 nm and

50nm. Again the graphs show that particle formation and particle number concentration are directly proportional to each other.

The data for the month of October which is a typical spring month is shown in Figure 5.11(a) for the particle diameters and Figure 5.11 (b) for the particle concentration.

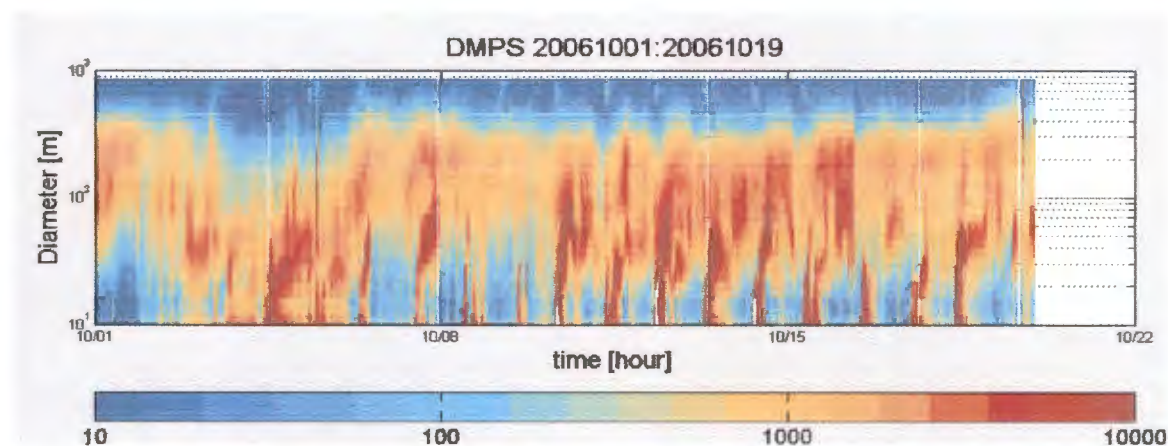


Figure 5.11: (a) Particle concentration against time for the October 2006

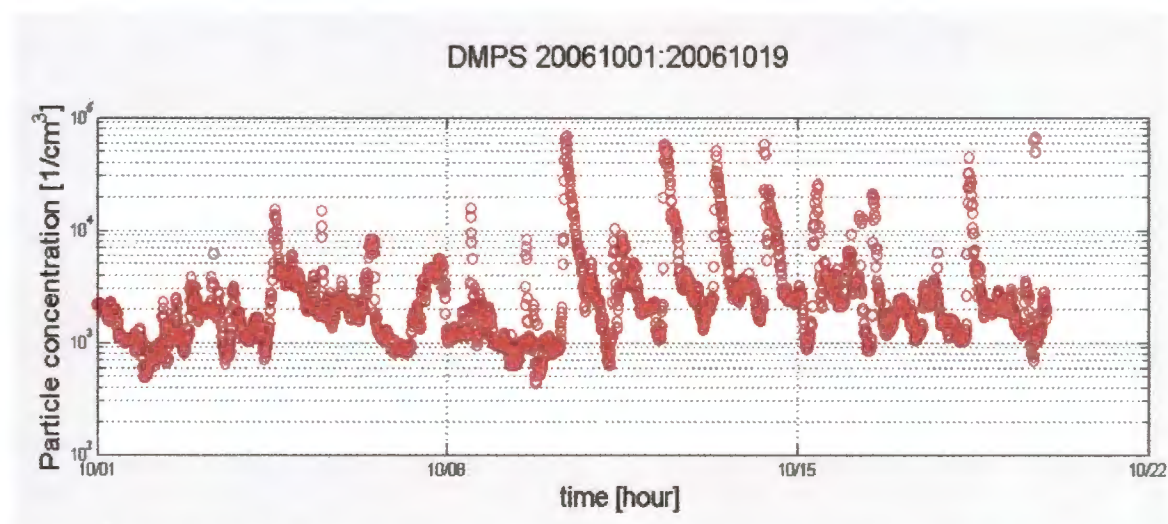


Figure 5.11: (b) Particle diameter and concentration against time for the October 2006

Figure 5.11 (a) shows that the formation rate was very low on the 2nd when compared to the formation rate on the other days of October. The particle

concentration increased from 600 cm^{-3} to 50000 cm^{-3} during the spring season. The particle diameter during the formation was between 10 and 70 nm. This may be due to global radiation, wind speed, wind direction and rain intensity which affected the particle formation during this season.

For a typical summer period February was chosen and the data for the period 11th to 19th February is shown in Figure 5.12 (a) and Figure 5.12 (b) for particle size and concentration respectively. The meteorological data in Figures 5.13.

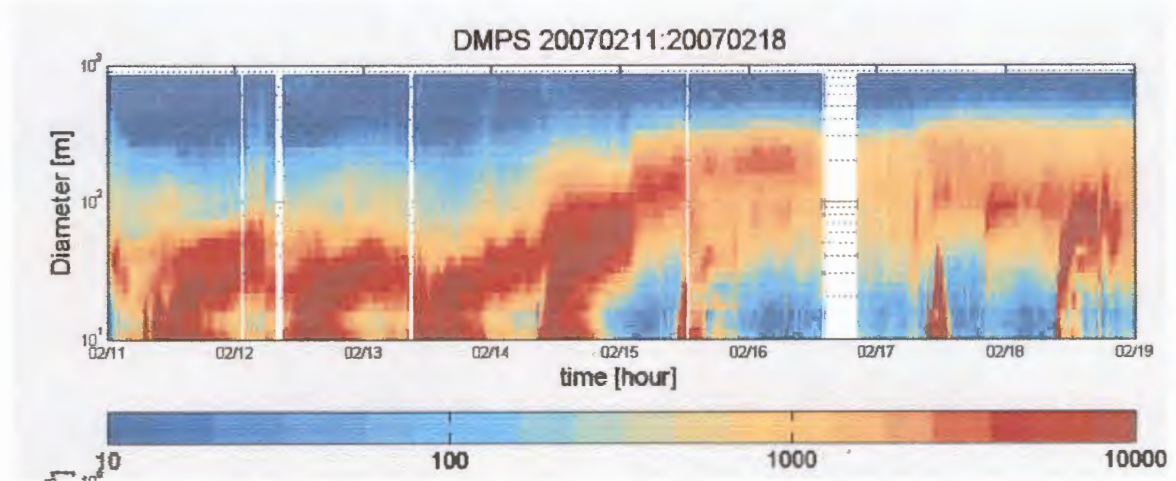


Figure 5.12: (a) Particle diameter against time for February 2007.

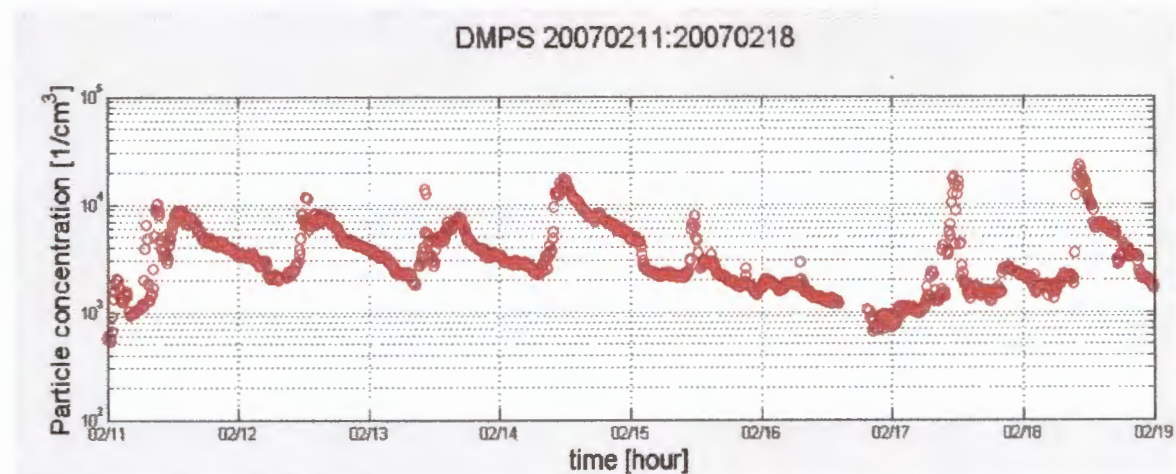


Figure 5.12:(b) Particle concentration against time for February 2007.

Figures 5.12 (a) and (b) show that particle formation and concentration on the 16th were lower than on other days such as the 14th. The particle concentration,

for instance, on the 16th was in the range 490 cm⁻³ to 8910 cm⁻³ and on the 14th it increased from 940 cm⁻³ to 1200 cm⁻³. Particle growth occurred between 10 nm and 50 nm in diameter. The global radiation, temperature, and wind speed were high and the relative humidity was low as shown in Figures 5.13. This clearly shows that, the formation and growth of particles is directly proportional to the particle concentration and wind speed.

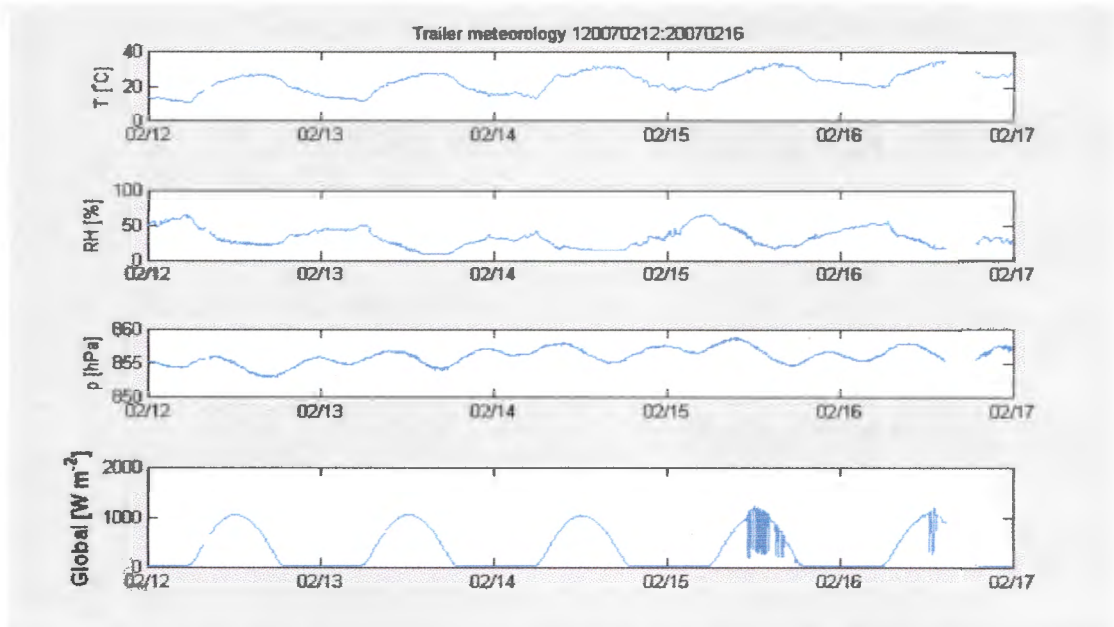


Figure 5.13 (a): The meteorological results from 12th to 16th of February 2007 for global radiation, relative humidity, pressure and temperature.

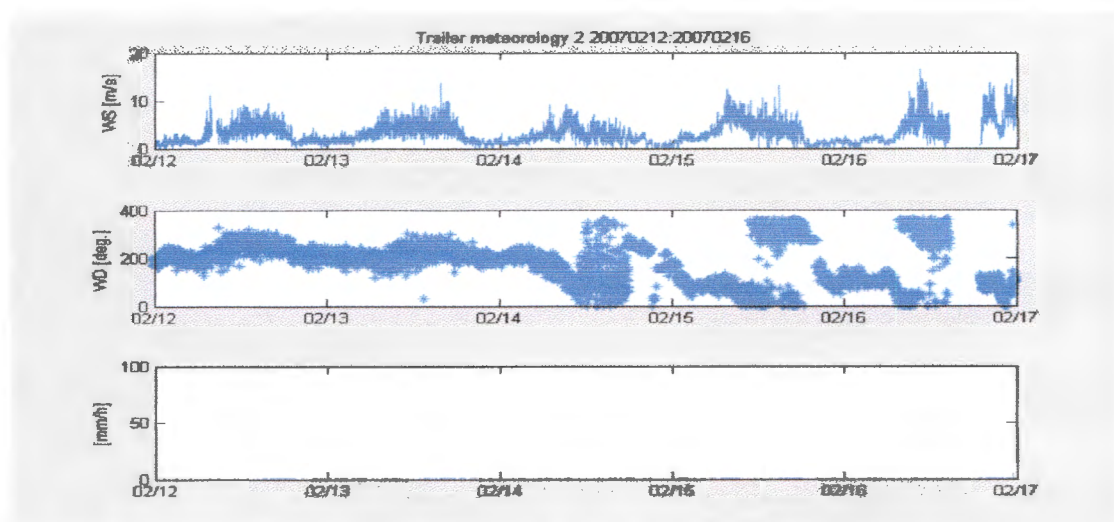


Figure 5.13 (b): The meteorological results from 12th to 16th of February 2007 for rain intensity, wind speed and direction.

Particle formation and growth for summer are shown in Figure 5.12, those for winter in Figure 5.10 and for spring in Figure 5.11. In winter the average concentration was less than 1000 cm^{-3} and it was more than 1000 cm^{-3} for summer. This is to be understood from the fact that concentration increases with temperature and global radiation which are higher in summer than in winter (Weingartner E, et al, 1999). In spring the particle concentration, particle formation and growth were high compared to those of summer and winter. This can be explained by noting that formation of new particles is affected by volatile organic compounds emitted by vegetation, and while the maximum is in summer, the frequency of the nucleation events peaks in spring and autumn (Kiendler-Scharr et al, 2009).

The relationship between particle growth and global radiation, temperature and wind speed is that particle growth is directly proportional to temperature, global radiation and wind speed, but it decreases as the humidity increases. The relative humidity was lower during event days, except for winter months. The reason may be that if relative humidity is high on cloudy days, there is less solar radiation to produce condensable vapours that cause the pre-existing aerosols

size to grow so that more surface area is available for vapour concentration (Boy and Kulmala, 2002). The temperature, on average, was higher on event days than non-event days, and this confirms the finding that most of the events occurred during the relatively warm season. Figure 5.14 shows the monthly variation of meteorological parameters taken over a period of one year from July 2006 to July 2007.

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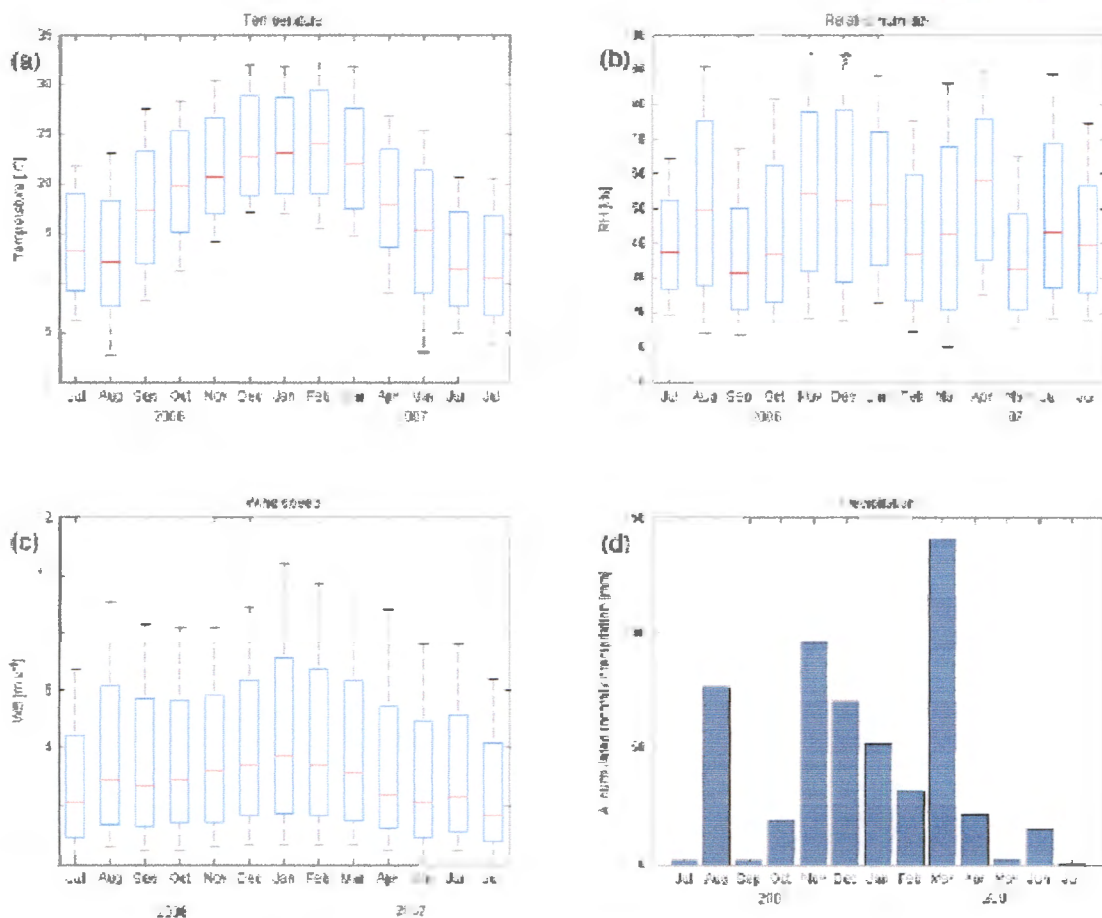


Figure 5.14: Variations of (a) temperature (b) relative humidity (c) wind speed (d) monthly accumulated precipitation. The red line in the middle of (a-c) is the 50% percentile.

5.2.VALUES FOR PARTICLE NUMBER CONCENTRATION GROWTH RATE AND FORMATION RATE

5.2.1. PARTICLE SIZE AND PARTICLE NUMBER CONCENTRATION

Figure 5.15 shows the relationship between particle size range and particle number concentration. This shows that the particle number concentration is directly proportional to the particle size range. Equation 4.1 was used to calculate the number concentration in Figure 5.15.

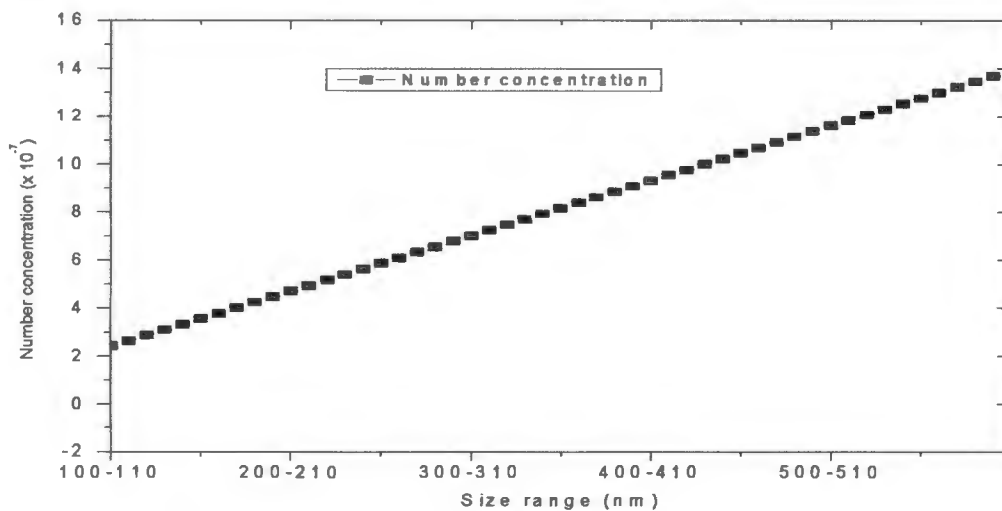


Figure 5.15: The relationship between size range and particle number concentration.

5.2.2 FORMATION VERSUS GROWTH RATE

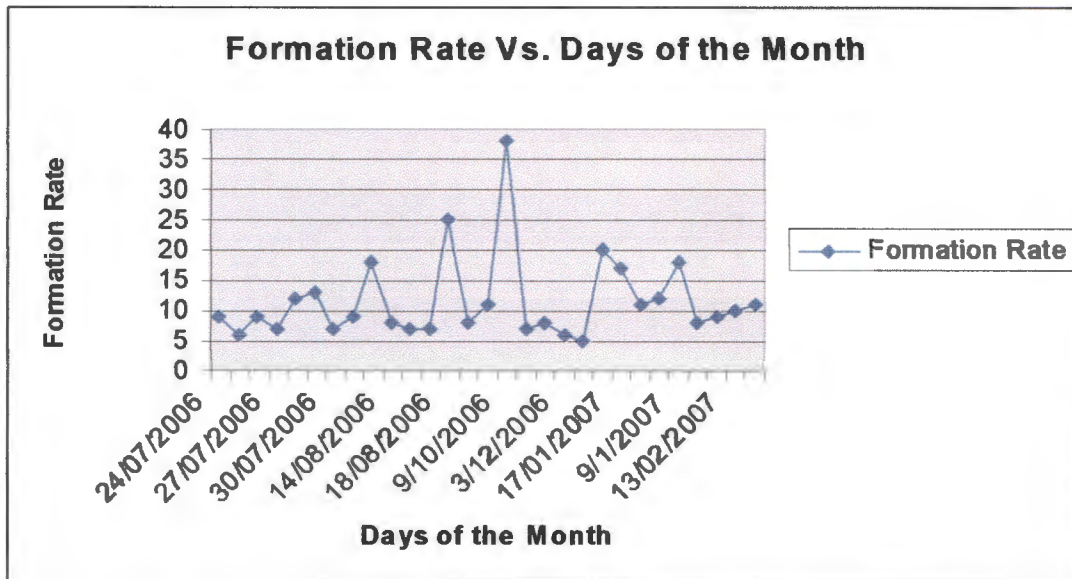


Figure 5.16: The relationship between formation rate and days of the month.

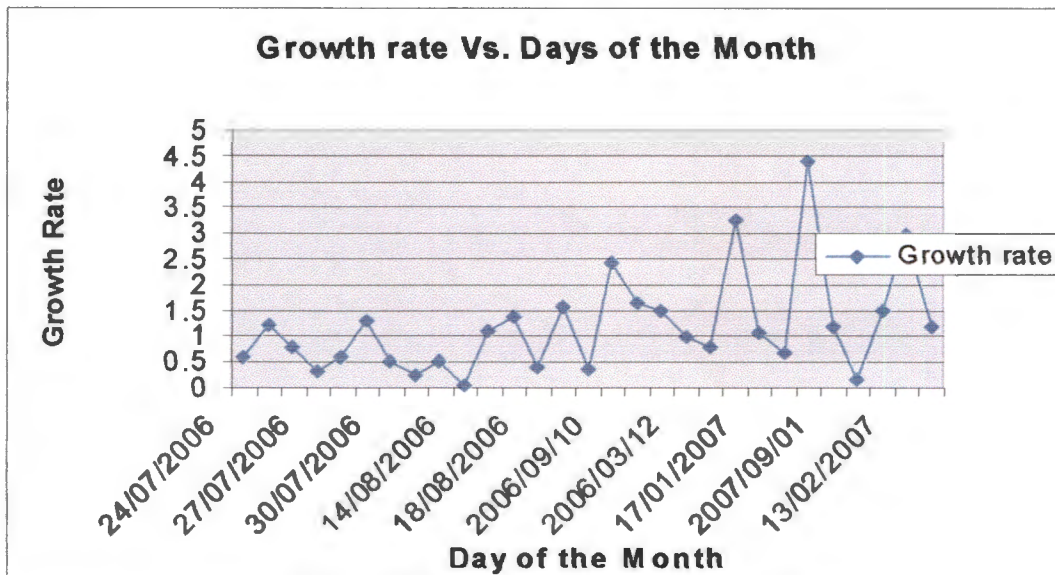


Figure 5.17: The relationship between growth rate and days of the month.

Figure 5.16 shows formation rate and Figure 5.17 growth rate with respect to different days of the month in different seasons. Equation 4.2 was used to calculate the growth rate and formation rate. The formation rate was measured in seconds because it takes a second for a particle to be formed and growth rate

was measured in hours because it normally takes an hour or more for a particle to grow. The particle formation rate was observed to be in the range 0.3 to 0.9 $\text{cm}^{-3}\text{s}^{-1}$ except in some days where it showed an unexpected range between 1.21 to 4 $\text{cm}^{-3}\text{s}^{-1}$, and the growth rate ranged from 6 to 12 nm/h. The formation and growth rate was found most of the time to be between 10 to 50 nm during this study. During the particle growth process, some of the particles were lost due to impaction, other particles were scavenged by the bigger ones, and some were lost during wet and dry deposition (Dal Maso et al, 2005). For the month of July, the growth rate and formation rate were negatively correlated but in summer and spring, the formation rate and growth rate were directly proportional to each other.

5.2.3. WIND DIRECTION AND PARTICLE CONCENTRATION

Wind roses summarize the occurrence of winds at a location, representing their strength, direction and frequency. Each directional branch on a wind rose represents wind originating from that direction. Figure 5.18 clearly shows the wind direction in which most of the particles originated. Most of particles were coming from Eastern sector (North-East, East and South East). This is the direction of the industrial and mining town of Rustenburg some 150 km away which was the most likely origin of anthropogenic pollution.

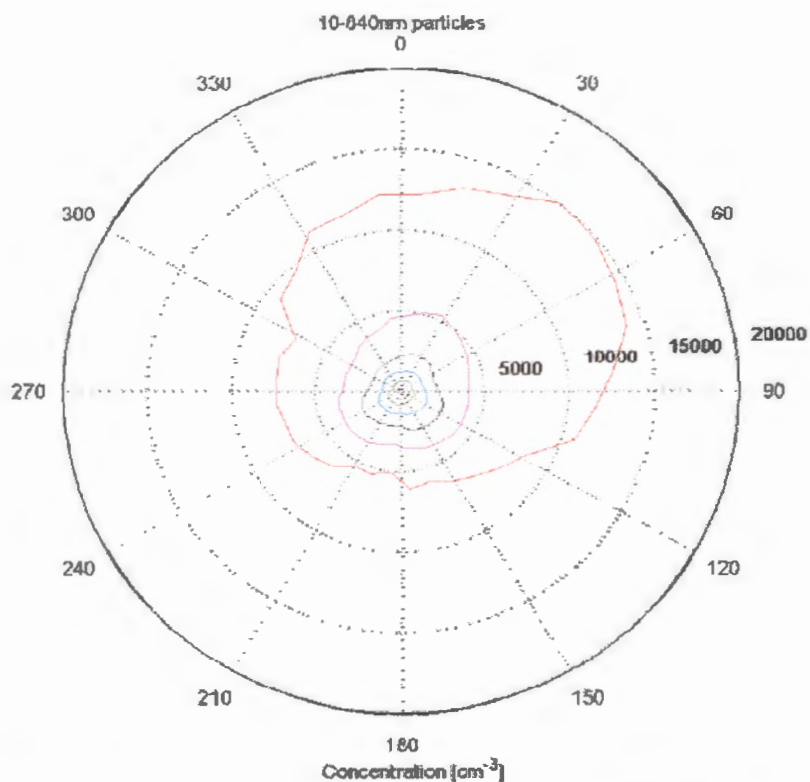


Figure 5.18: Total number concentration of particles as a function of wind direction. Different colors represent percentiles: green: 5%, blue 25%, black 50%, magenta 75% and red 95%.

CHAPTER 6

CONCLUSIONS

The number size distribution of particles with radii in the range 0.01 to 0.84 μm was measured in the Botsalano game reserve, and the occurrences of new particle formation as well as their growth rates were also determined. The measurements were carried out daily and continuously from July 2006 to July 2007. Typical number concentrations ranged from 100 cm^{-3} to 1000 cm^{-3} . During aerosol generation some particles were scavenged by other particles. Particle losses due to coagulation and dry deposition were also observed. Despite some differences in the results obtained, evidence of particle growth was obtained. An observed relationship between particle size and particle number concentration is that they are directly proportional to each other. There is a positive correlation between particle size and concentration because when size increases the concentration increases as well as shown in Fig 5,15.

Average and total number concentration for every week were found to be 700 cm^{-3} and 10000 cm^{-3} . Daily averages varied between 740 cm^{-3} and 10000 cm^{-3} . There was not much difference in particle formation and growth rates for weekends and weekdays, they only depended on the activity that happened on that particular day. Seasonal variations of total number concentration were also been observed, and the particle formation rates were found to be highest during spring with a mean value of $4.4\text{ cm}^{-3}\text{s}^{-1}$. During other seasons the mean formation rates ranged from 1.2 to $4\text{ cm}^{-3}\text{s}^{-1}$. The mean growth rates varied from 6 to 12 nm/h with high values occurring in spring and summer and low values in winter. The low values of formation rates in winter can be attributed to generally lower temperatures as a result of reduced global radiation. The onset of spring raises both temperature and the terpene emissions by vegetation and these result in higher growth rates. Starting times of the formation events were affected by relative humidity, wind speed and wind direction for different seasons.

One of the main objectives of the study was to determine the relationship between particle formation and particle growth, and the results obtained show that these are directly proportional to each other. The results obtained such as Figures 5.3 (a) and 5.4 (a) show that particle growth takes place mostly during the day around 12 hour, a little before in summer and a little after in winter. The average particle growth was also shown to depend on the external global radiation. This can be seen from a comparison of graph of particle growth versus time in Figure 5.1 (a) and that of temperature and global radiation as functions of time in Figure 5.2 (a). Many factors that influence growth such as the terpene emissions by vegetation are important at warmer temperatures such as those occurring in spring and summer. The variations in the observed graphs can be attributed to natural sources and socio-economic activities in the area. The sizes of the various peaks may differ, depending on the season and ambient temperature. Because socio-economic activities are performed mostly during the day, a gradual increase of particle formation and growth can be observed during the course of the day.

Meteorological conditions can favour or suppress particle formation. Higher solar radiation promotes the photochemical oxidation processes, for example, through increasing the number of OH radicals. Radiation also increases the particle formation and growth (Kulmala et al., 2001). Increased relative humidity has been linked with suppressed particle formation due to the wet scavenging of particles and precursor gases and is increased by growing particles (Hamed et al., 2007). Meteorological conditions during spring were favorable for particle formation. The global radiation was most intense during the spring (mean value of 203.4 Wm^{-2}). Spring was also relatively dry allowing the particles and the precursor gases to not be scavenged by wet deposition.



The number size distribution of particles of radii 0.01 to 0.84 μm has been measured and new particle formation events were observed in the Savannah environment. It was found that particle formation and growth were positively

correlated, and the influence of meteorological parameters was observed. It is suggested here that a further development of the project would be to carry out a chemical analysis of the aerosols in the area and determine their concentrations on a seasonal basis as this information would enable the determination of the influence of the various gaseous compounds on particle formation.

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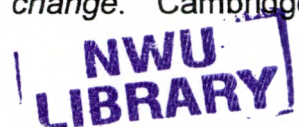
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APPENDIX A

TABLES

Table A (0.1): The table that shows the flow rate and tube diameter combinations with Reynolds number 2000

Flow rate (LPM)	Pipe diameter (cm)
6	0.4
12	0.8
16.67	1.2
30	2.1
100	7
143	10
220	15
286	20

Table A (0.2): The table shows that size range and number concentration

Size Range (nm)	$\frac{dN}{dLog(dp)}$
10-20	3.32×10^{-9}
20-30	5.678832×10^{-8}
30-40	8.0×10^{-7}
40-50	1.019×10^{-7}
50-60	1.2629×10^{-7}
60-70	1.4937×10^{-7}
70-80	1.7244×10^{-7}
80-90	321.9549×10^{-7}
90-100	2.1854×10^{-7}

100-110	2.4159×10^{-7}
110-120	2.6463×10^{-7}
120-130	2.8767×10^{-7}
130-140	3.1071×10^{-7}
140-150	3.3374×10^{-7}
150-160	3.5678×10^{-7}
160-170	3.7981×10^{-7}
170-180	4.0284×10^{-7}
180-190	4.2587×10^{-7}
190-200	4.4891×10^{-7}
200-210	4.7194×10^{-7}
210-220	4.9497×10^{-7}
220-230	5.1800×10^{-7}
230-240	5.4103×10^{-7}
240-250	5.6406×10^{-7}
250-260	5.8708×10^{-7}
260-270	6.1011×10^{-7}
270-280	6.3314×10^{-7}
280-290	6.5617×10^{-7}
290-300	6.7920×10^{-7}
300-310	7.0223×10^{-7}
310-320	7.2525×10^{-7}
320-330	7.4828×10^{-7}
330-340	7.7131×10^{-7}
340-350	7.9434×10^{-7}
350-360	8.1736×10^{-7}
360-370	8.4039×10^{-7}
370-380	8.6342×10^{-7}
380-390	8.8645×10^{-7}
390-400	9.0947×10^{-7}

400-410	9.3250×10^{-7}
410-420	9.5553×10^{-7}
420-430	9.7855×10^{-7}
430-440	1.0016×10^{-6}
440-450	1.0246×10^{-6}
450-460	1.0476×10^{-6}
460-470	1.0707×10^{-6}
470-480	1.0937×10^{-6}
480-490	1.1167×10^{-6}
490-500	1.1397×10^{-6}
500-510	1.1628×10^{-6}
510-520	1.1858×10^{-6}
520-530	1.2088×10^{-6}
530-540	1.2318×10^{-6}
540-550	1.2549×10^{-6}
550-560	1.2779×10^{-6}
560-570	1.3009×10^{-6}
570-580	1.3240×10^{-6}
580-590	1.3470×10^{-6}
590-600	1.3700×10^{-6}

Table A (0.3): The table shows size range N_p particle concentration, the normalized concentration

Size range ΔD_p (μm)	Concentration N_p (cm^{-3})	Normalized Concentration N_{norm} ($\mu\text{m}^{-1} \cdot \text{cm}^{-3}$)
0.001 – 0.01	100	11111
0.01 – 0.02	200	20000
0.02 – 0.03	30	3000
0.03 – 0.04	20	200
0.04 – 0.08	40	100
0.08 – 0.16	60	750
0.16 – 0.32	200	1250
0.32 – 0.64	180	563
0.64 – 1.25	60	98
1.25 – 2.5	20	16
2.5 – 5.0	5	2
5.0 – 10.0	1	0.2

Table A (0.4): Table a (4) shows the particles formation rate and concentration range for a particular day within 10 to 40 nanometer diameter.

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Date	Dp Range (nm)	Concentration Range (cm^{-3})	Formation rate ($\text{cm}^{-3} \cdot \text{s}^{-1}$)	Growth rate nm/h
24-07-2006	10 - 40	260-7120	0.5678	9
25-07-2006	10 - 30	390-11920	1.2167	6
26-07-2006	10 - 30	600-3530	0.7792	9
27-07-2006	10-20	180-1780	0.3253	7
28-07-2006	10-20	130-1780	0.5874	12
29-07-2006	10-50	208-8560	1.2778	13
30-07-2006	10-20	500-1680	0.5268	7
12-08-2006	10-20	20-320	0.2377	29
13-08-2006	10-40	190-2360	0.5210	18
14-08-2006	10-30	80-240	0.0257	8
16-08-2006	10-30	280-4370	1.0987	7

17-08-2006	10-30	300-5290	1.3588	7
18-08-2006	10-40	250-3120	0.4062	25
10-09-2006	10-40	270-13130	1.5635	8
07-10-2006	10-40	100-2000	0.3327	11
09-10-2006	10-30	140-6470	2.4266	38
11-10-2006	10-30	190-6990	1.6341	7
02-12-2006	10-50	490-8910	1.4736	8
03-12-2006	10-30	590-9852	0.9645	6
04-12-2006	10-30	210-4570	0.7906	5
05-12-2006	10-50	250-5680	3.25	20
17-01-2007	10-50	300-5270	1.0702	17
18-01-2007	10-40	180-4520	0.6493	11
08-01-2007	10-40	1580-66420	4.3886	12
09-01-2007	10-40	150-2890	1.1811	18
11-01-2007	10-20	130-620	0.1482	8
12-01-2007	10-30	2550-9460	1.4879	9
13-02-2007	10-50	2550-12050	2.9773	10
14-02-2007	10-60	2090-15210	1.1891	11

APPENDIX B

METEOROLOGICAL RESULTS

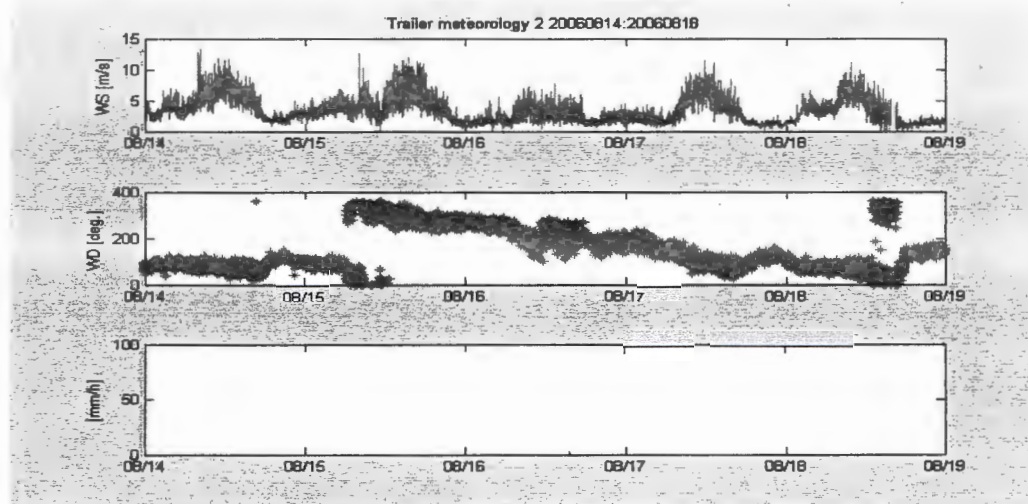


Figure B(1) The meteorological results from 14th to 18th of August 2006 for meteorological results for Rain intensity, wind speed and direction.

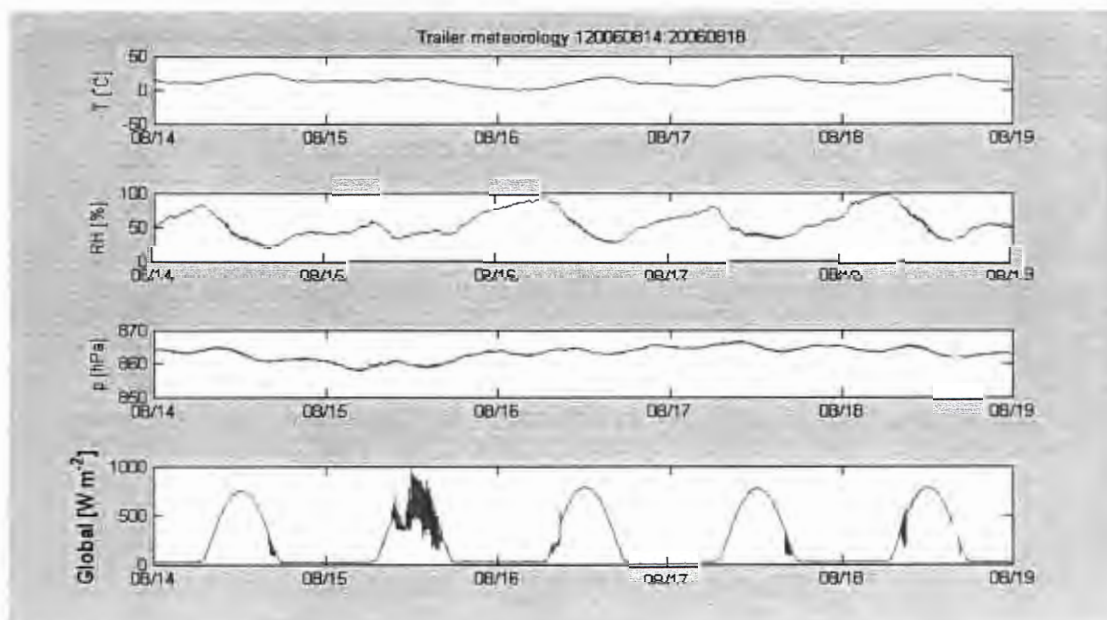


Figure B (2) The meteorological results from 14th to 18th of August 2006 for Global radiation, relative humidity, pressure and temperature.

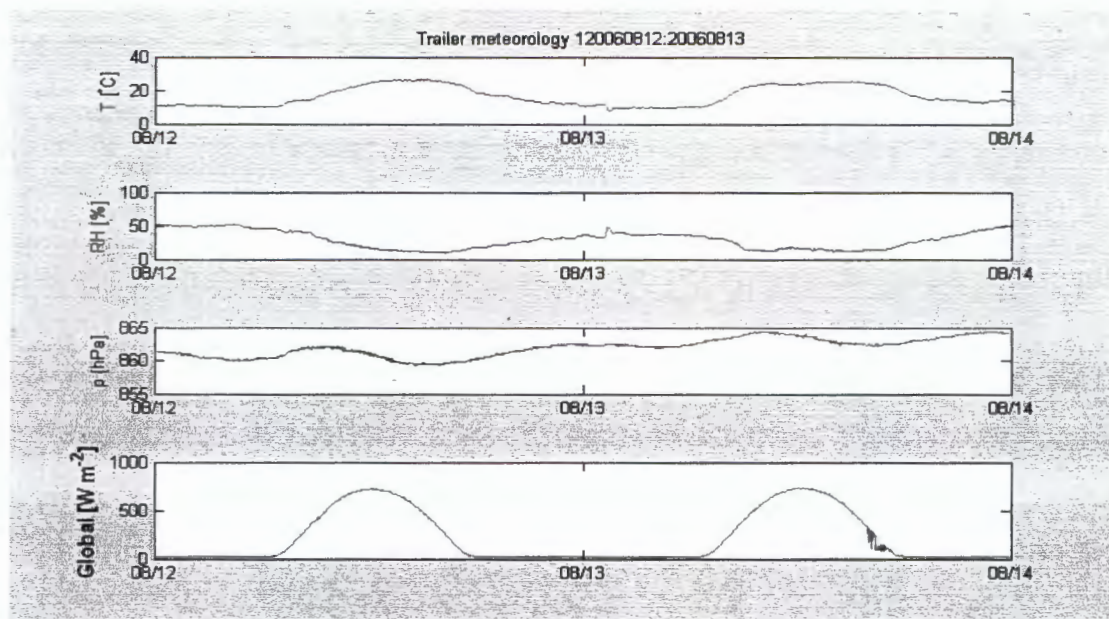


Figure B (3) Weekend meteorological results for Global radiation, relative humidity, pressure and temperature for the weekend of the 12th and 13th of August 2006.

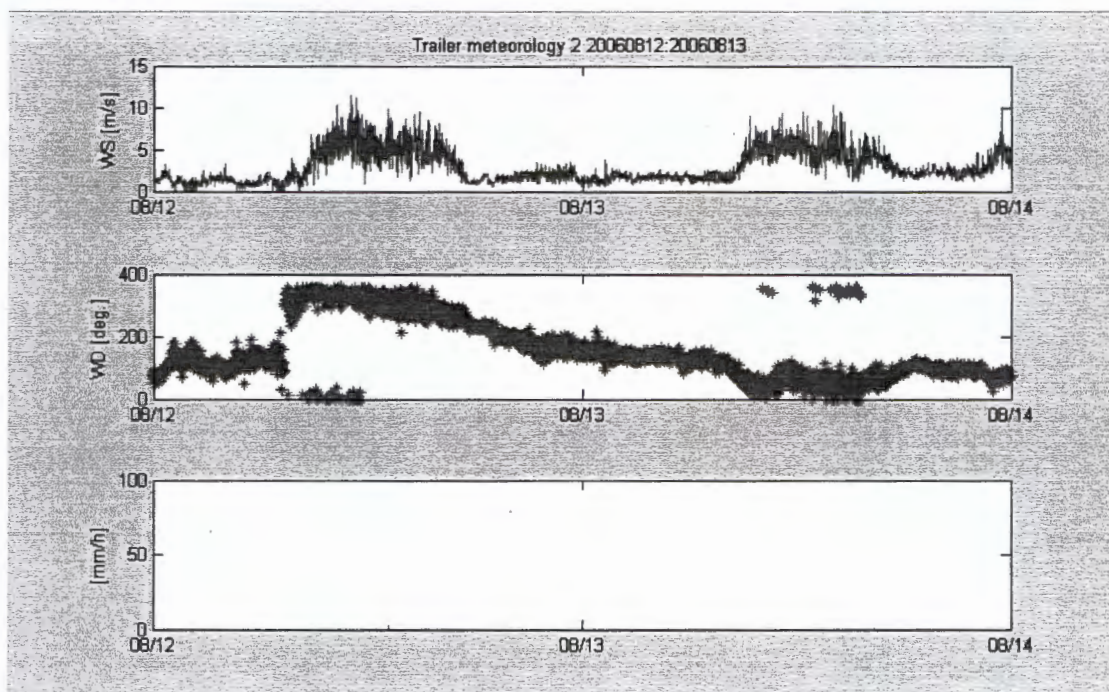


Figure B (4) The meteorological results for Rain intensity, wind speed and direction for the weekend of the 12th and 13th of August 2006.

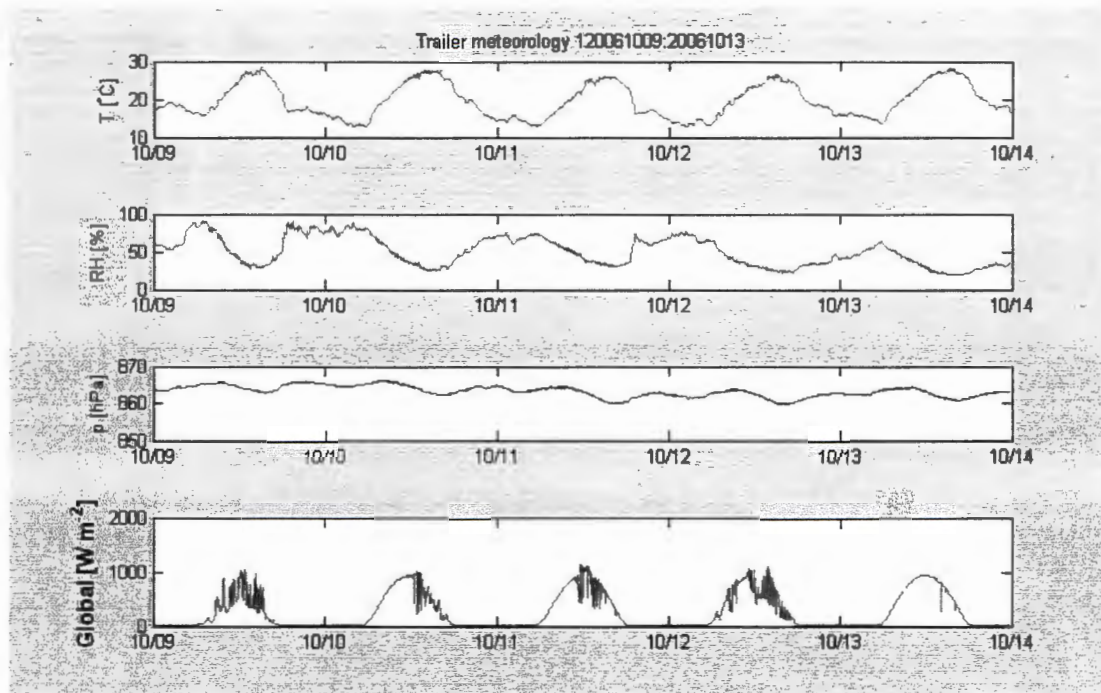


Figure B (5) The meteorological results from 9th to 14th of October 2006 for Global radiation, relative humidity, pressure and temperature.

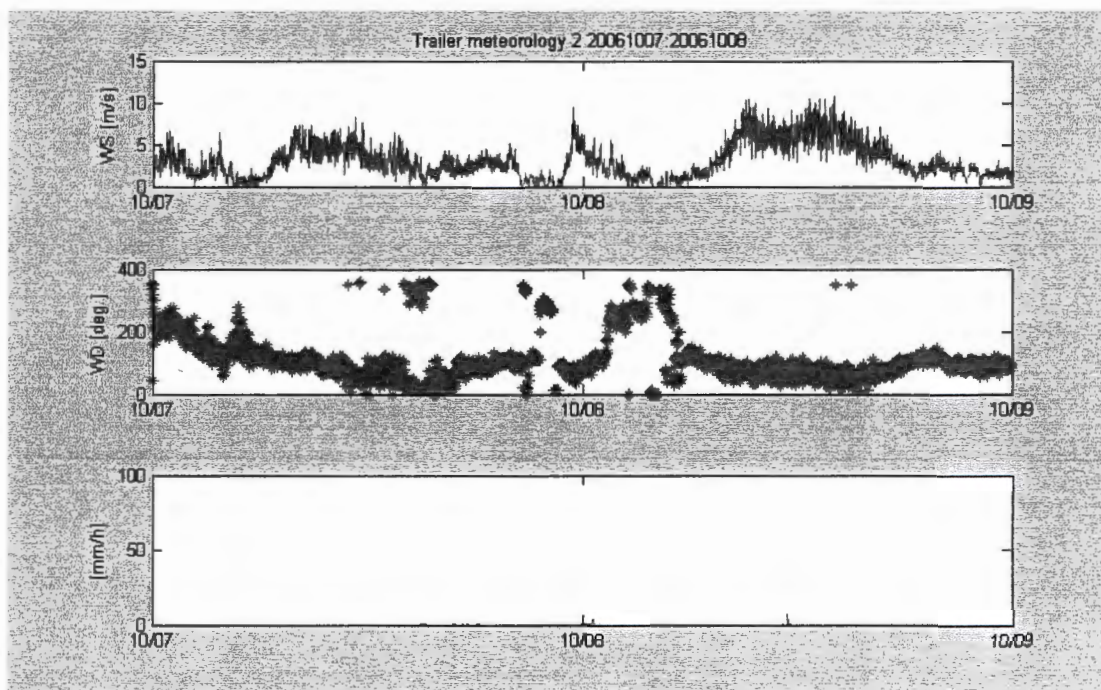


Figure B(6) The meteorological results from 7th to 8th of October 2006 for meteorological results for Rain intensity, wind speed and direction.

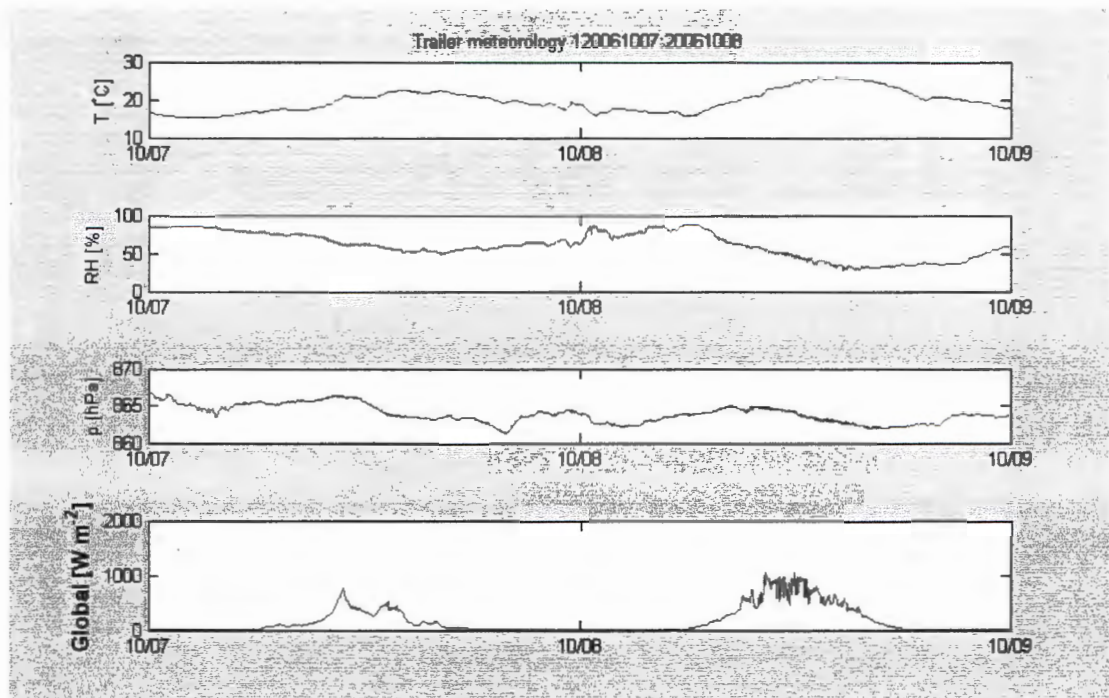


Figure B (7)The meteorological results for Global radiation, relative humidity, pressure and temperature for the weekend of the 7th and 8th of October 2006.

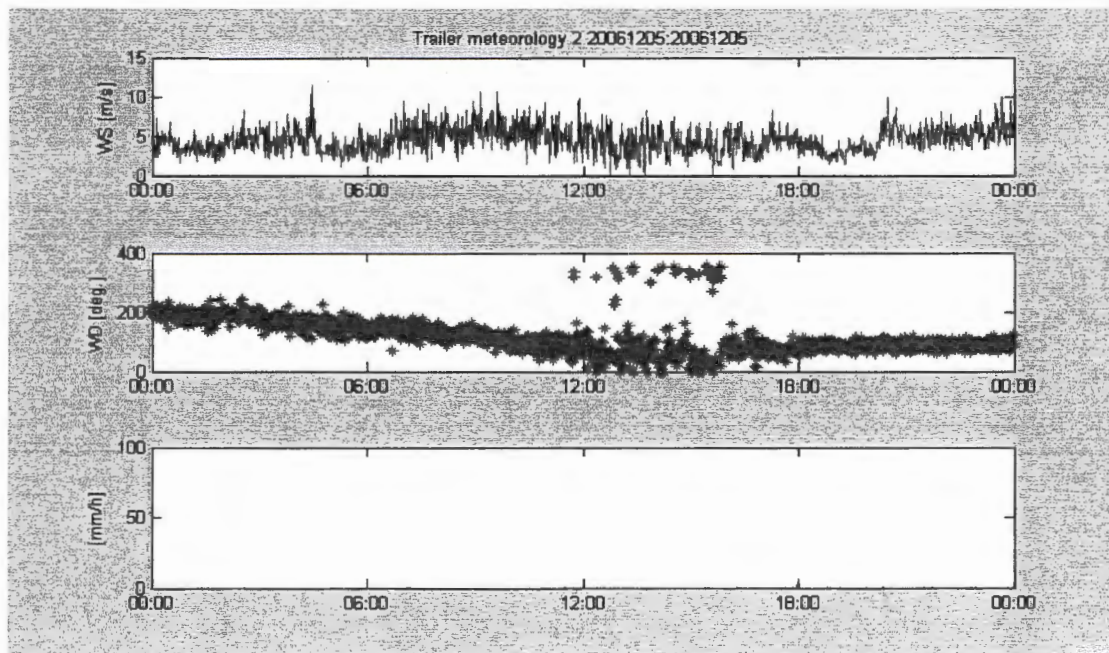


Figure B(8) The meteorological results for 5th of December 2006 for meteorological results of Rain intensity, wind speed and direction.

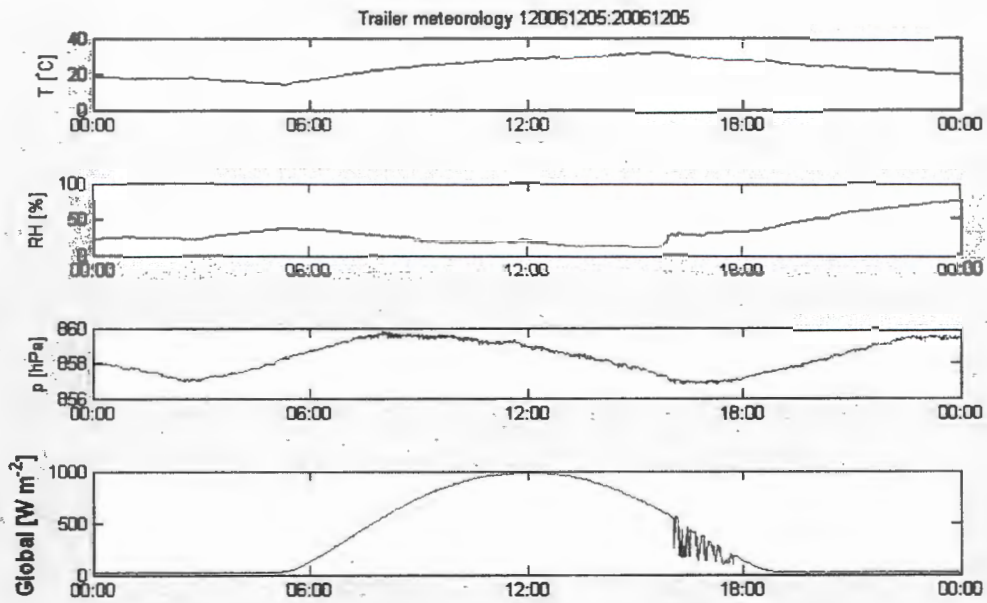


Figure B (9)The meteorological results for 5th of December 2006 for meteorological results of Global radiation, relative humidity, pressure and temperature.

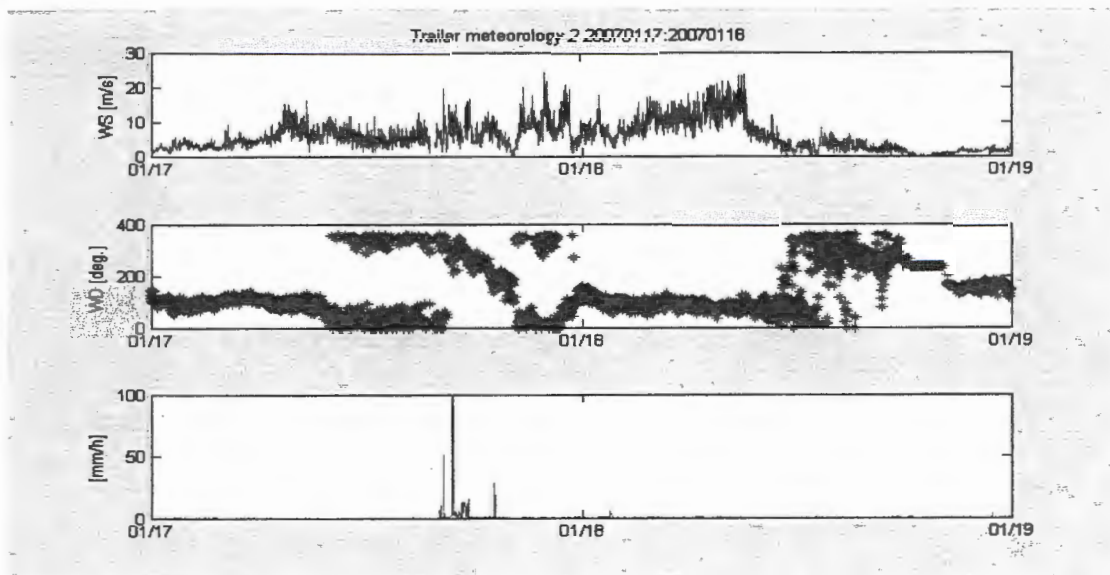


Figure B (10)The meteorological results from 17th to 18th of January 2007 for meteorological results for Rain intensity, wind speed and direction.

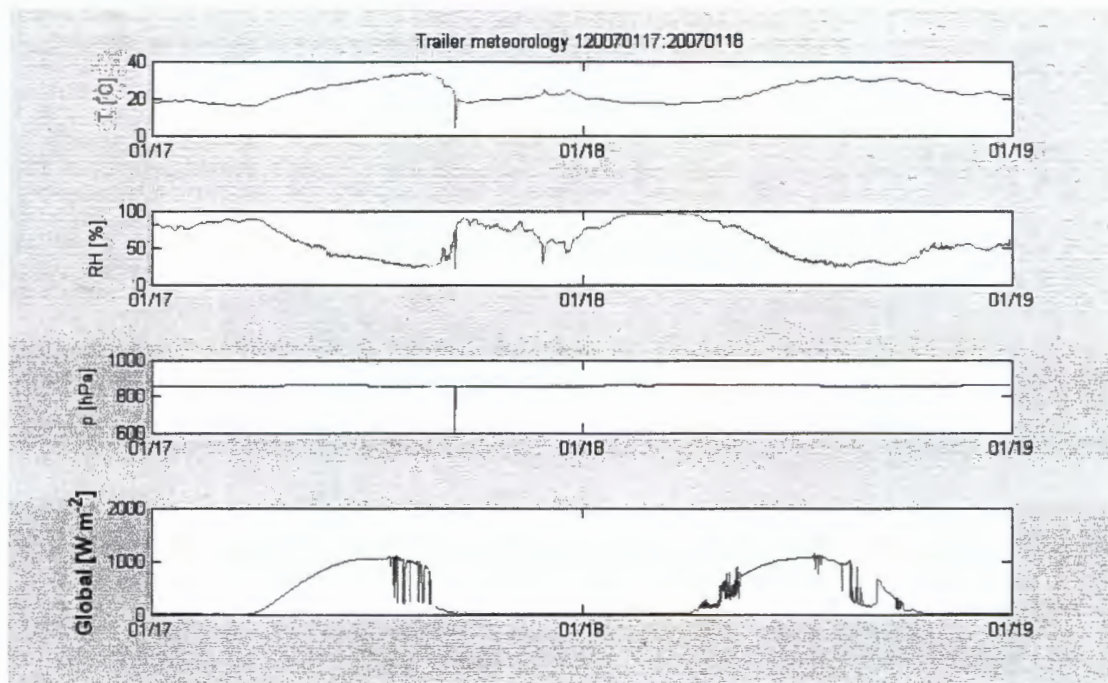


Figure B (11) The meteorological results for Global radiation, relative humidity, pressure and temperature for the weekend of the 17th and 18th of January 2007.

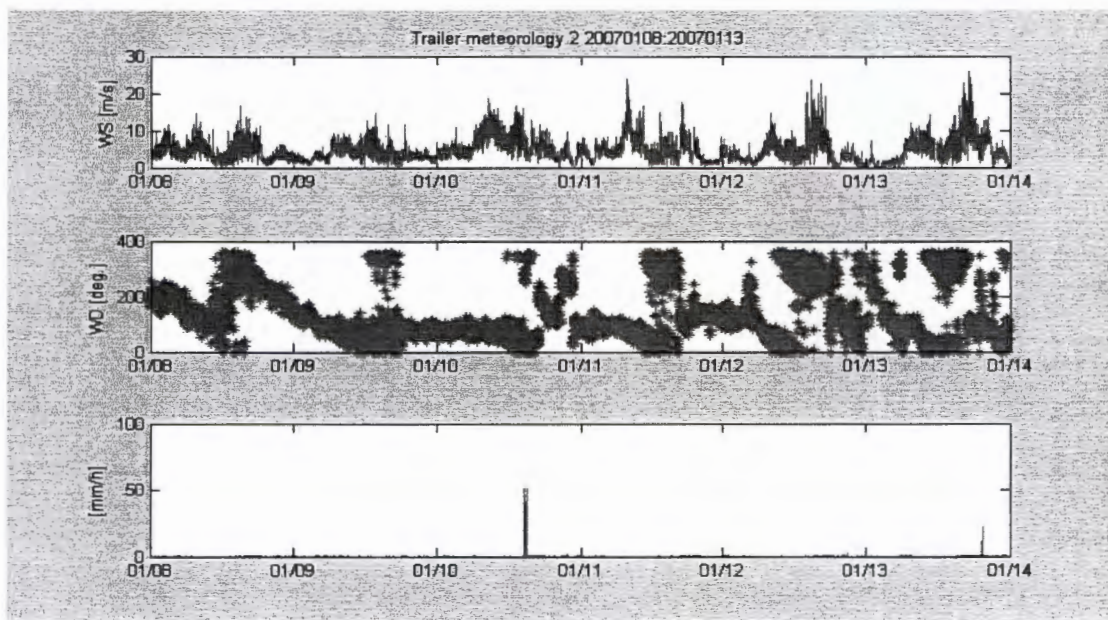


Figure B (12) The meteorological results from 8th to 13th of August 2006 for meteorological results for Rain intensity, wind speed and direction from Monday to Friday.

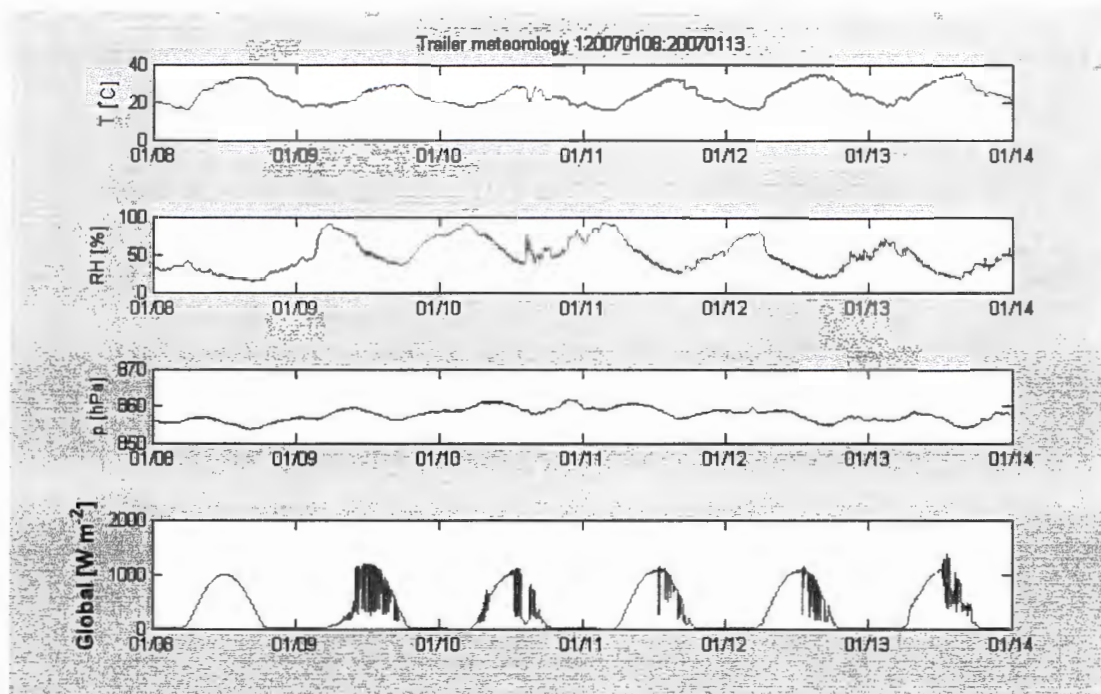


Figure B (13)The meteorological results from 8th to 13th of January 2007 for Global radiation, relative humidity, pressure and temperature from Monday to Friday.

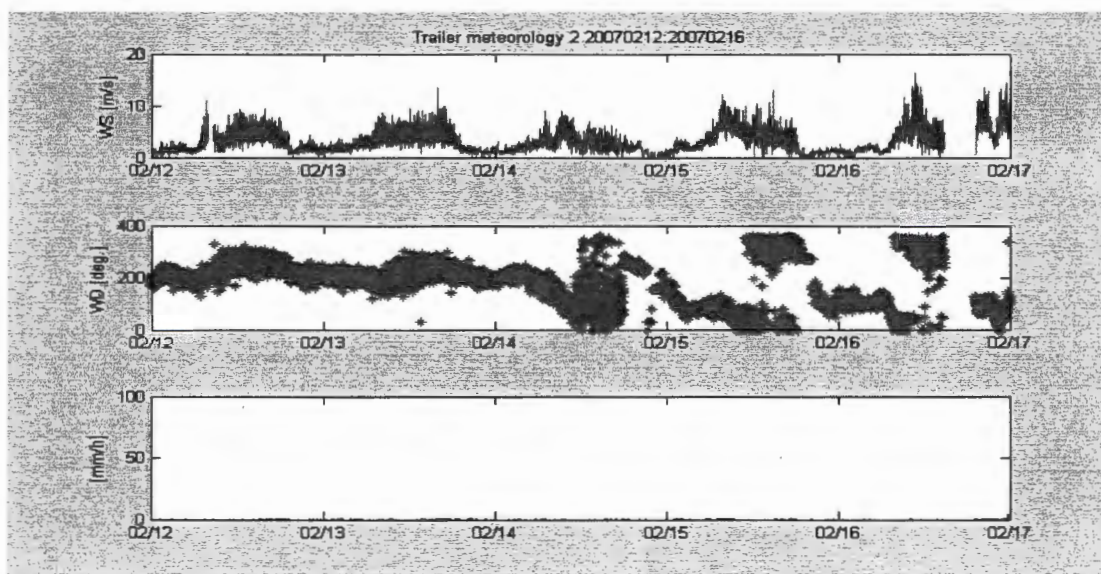


Figure B (14)The meteorological results from 12th to 16th of February 2007 for meteorological results for Rain intensity, wind speed and direction.

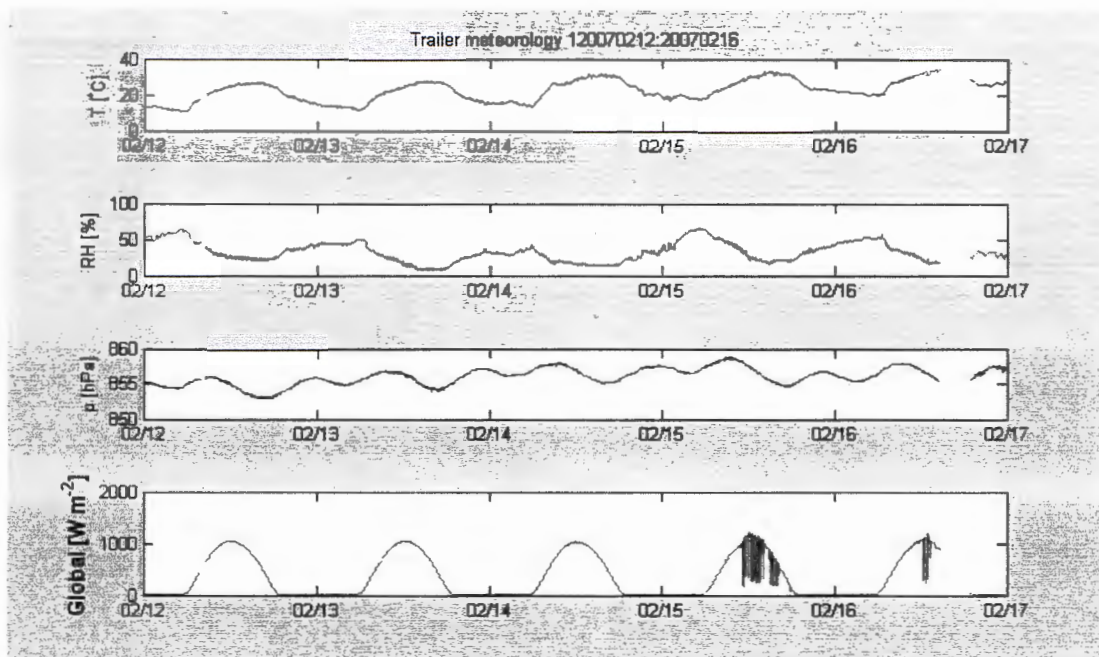


Figure B (15) The meteorological results from 12th to 16th of February 2007 for Global radiation, relative humidity, pressure and temperature.

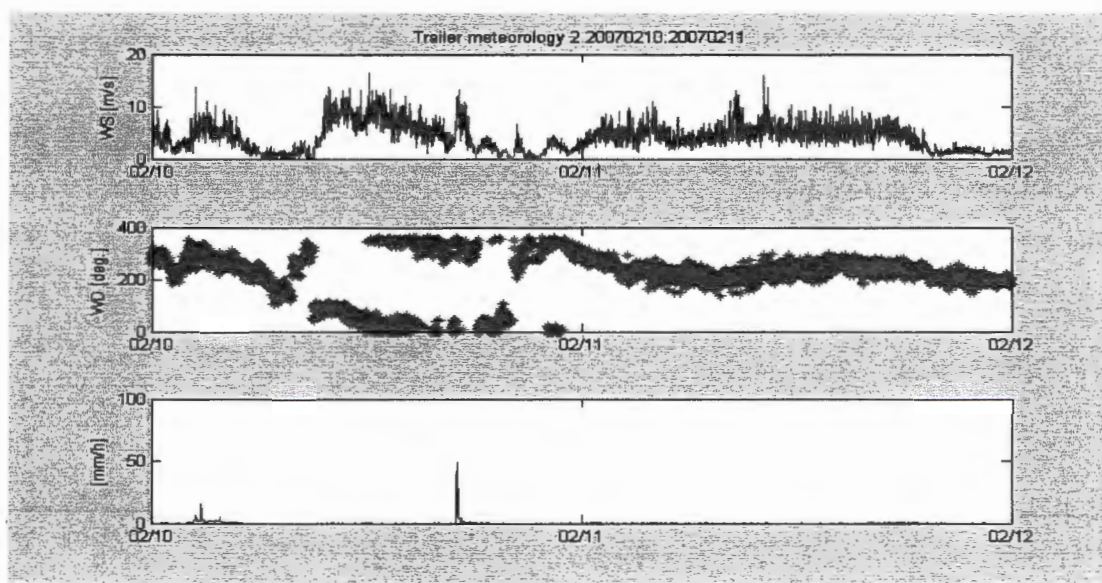


Figure B (16) The meteorological results from 10th to 11th of February 2007 for meteorological results for Rain intensity, wind speed and direction.

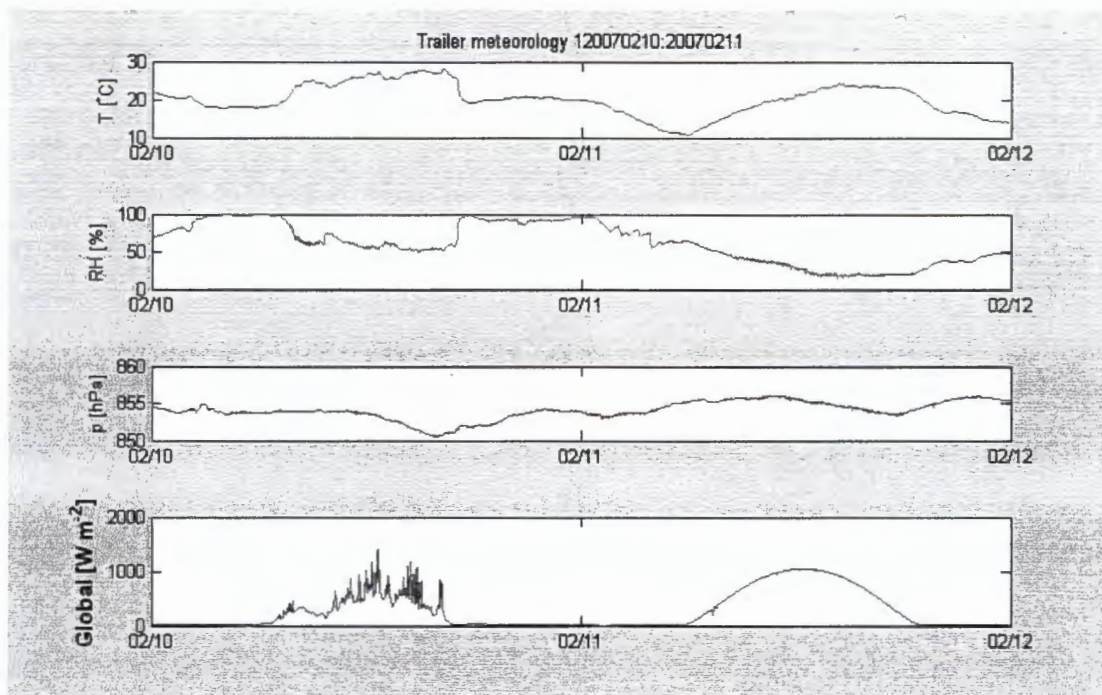


Figure B (17)The meteorological results for Global radiation, relative humidity, pressure and temperature for the weekend of the 10th and 11th of February 2007.

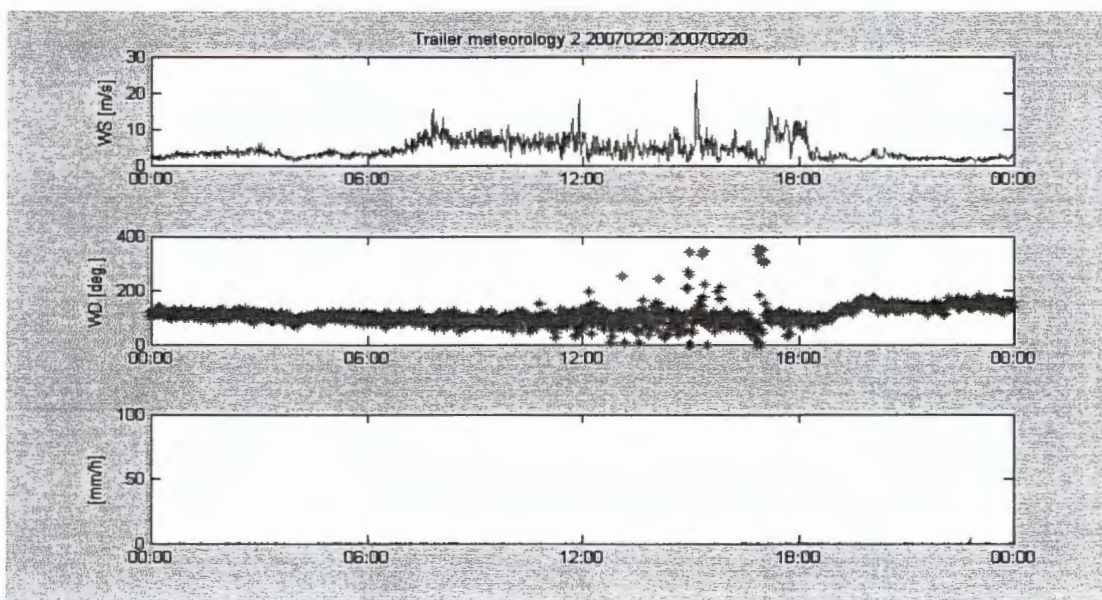


Figure B (18)The meteorological results for 20th of February 2007 for meteorological results of Rain intensity, wind speed and direction.

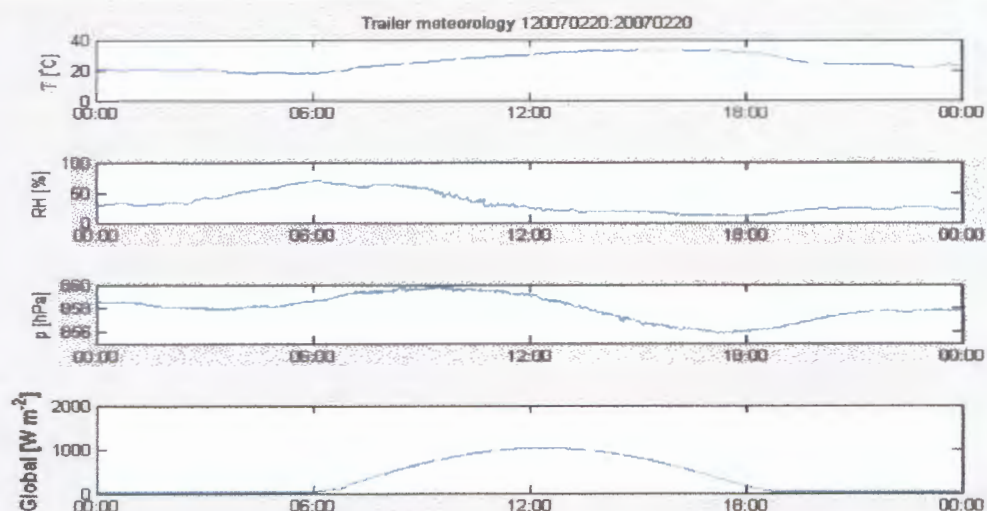


Figure B (19) The meteorological results for 20th of February 2007 for meteorological results of Global radiation, relative humidity, pressure and temperature.

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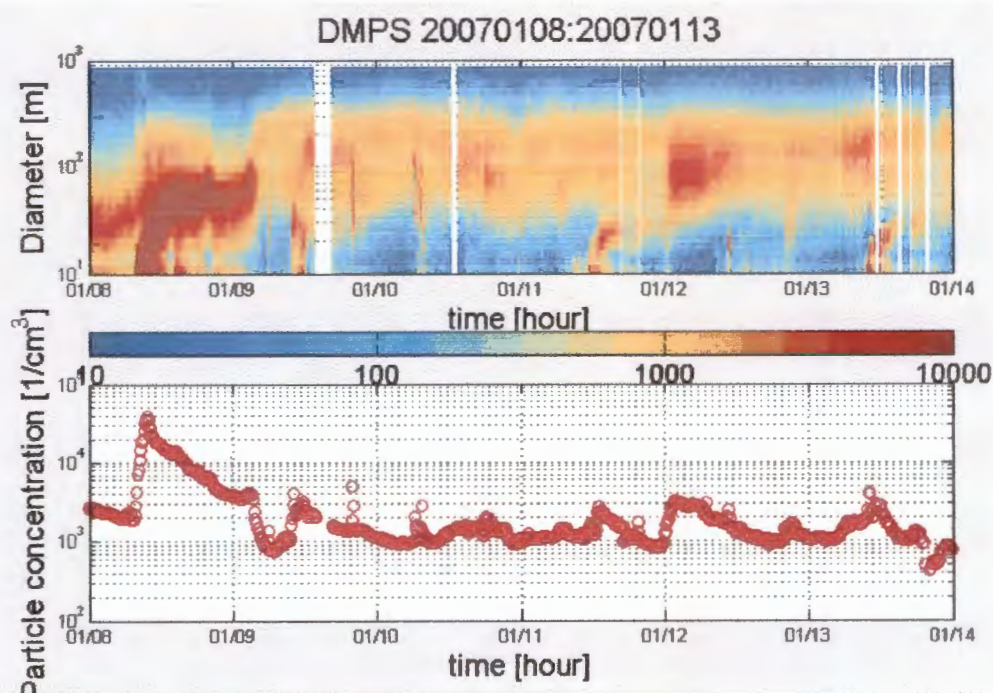


Figure B (20) Particle concentration and diameter against time for 8th of January to the 14th January 2007

APPENDIX C

PROGRAM USED FOR DATA ANALYSIS

Particles and particles concentration

```
function plot_particles(numconc, dp_part, beg, endi)
% LL 18.1.2007
% input: particle dndlogdp [cm-3]
% particle diameters in [nm]

date1=[datestr(numconc(1,1),'yyyy') datestr(numconc(1,1),'mm') datestr(numconc(1,1),'dd')];
date2=[datestr(numconc(end,1),'yyyy') datestr(numconc(end,1),'mm')
datestr(numconc(end,1),'dd')];

figure
set(gcf,'papertype','a4letter')
orient tall

subplot(2,1,2)

semilogy(numconc(:,1), numconc(:,2), 'or'); % plot total particle concentration between 10 and 840
nm.

set(gca,'ylabel',text(0,0,'Particle concentration [1/cm^3]'))
set(gca,'xlabel',text(0,0,'time [hour]'))
set(get(gca,'xlabel'),'Fontname','helvetica','fontsize',16)
set(get(gca,'ylabel'),'Fontname','helvetica','fontsize',16)
grid
axis([beg endi+1 100 1e5])
%datetick(gca,'x','dd')
datetick

subplot(2,1,1)

Zdata=real(log10(numconc(:,3:end)+1e-6));
```

```
pcolor(numconc(:,1),dp_part,Zdata'); % surface plot of DMPS-data
```

```
colormap(jet(250));
```

```
caxis([1 4]);
```

```
set(gca,'yscale','log')
```

```
set(gca,'Ylim',[10 1000])
```

```
grid
```

```
%datetick(gca,'x','dd')
```

```
datetick
```

```
shadingflat
```

```
set(gca,'ylabel',text(0,0,'Diameter [nm]'))
```

```
set(gca,'xlabel',text(0,0,'time [hour]'))
```

```
set(get(gca,'xlabel'),'Fontname','helvetica','fontsize',16)
```

```
set(get(gca,'ylabel'),'Fontname','helvetica','fontsize',16)
```

```
set(gca,'title',text(0,0,['DMPS ' date1 ':' date2']))
```

```
set(get(gca,'title'),'Fontname','helvetica','fontsize',18)
```

```
% make a colorbar
```

```
gcc=colorbar('horiz');
```

```
set(gcc,'xlim',[1 4],'xtick',[1 2 3 4],'fontsize',14)
```

```
set(gcc,'xticklabel',[10,100,1000,10000])
```

```
V=get(gcc,'position');
```

```
set(gcc,'position',[V(1) V(2)-0.1 V(3) V(4)])
```

```
disp('DMPS plot finished!')
```

Metheorological

```
function plot_meteo(meteoro,beg,endi)
```

```
% LL 16.2.2007
```

```
%plots meteorology
```

```
%
```

```
date1=[datestr(meteoro(1,1),'yyyy') datestr(meteoro(1,1),'mm') datestr(meteoro(1,1),'dd')];
```

```
date2=[datestr(meteoro(end,1),'yyyy') datestr(meteoro(end,1),'mm') datestr(meteoro(end,1),'dd')];
```

```

% Columns:
%1: time [matlabs format]
%2: PAR-radiation [umol/s] => converted to global radiation [W/m2] by multiplying with 500
%3: Temperature [C]
%4: RH [%]
%5: Wind speed
%6: Wind direction
%7: precipitation [mA]. Converted to [mm/h] with program "reinconv.m"
%8: pressure [hPa]

```

```
figure
```

```
subplot(4,1,1)
```

```
plot(meteoro(:,1),meteoro(:,3))
```

```
hold on
```

```
ylabel('T [^\circ C]')
```

```
a=axis;
```

```
axis([beg endi+1 a(3) a(4)]);
```

```
%datetick(gca,'x','dd')
```

```
datetick
```

```
grid
```

```
set(gca,'title',text(0,0,['Trailer meteorology 1' date1 ':' date2]))
```

```
subplot(4,1,2)
```

```
plot(meteoro(:,1),meteoro(:,4))
```

```
hold on
```

```
ylabel('RH [%]')
```

```
a=axis;
```

```
axis([beg endi+1 a(3) a(4)]);
```

```
%datetick(gca,'x','dd')
```

```
datetick
```

```
grid
```

```
subplot(4,1,3)
```

```
plot(meteoro(:,1),meteoro(:,8))
```