

# UNIVERSITY OF JOHANNESBURG



## **STEAM EXTRACTION OF ESSENTIAL OILS: INVESTIGATION OF PROCESS PARAMETERS**

**By**

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## **Declaration by candidate**

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I hereby declare that this dissertation is my own, unaided work. It is being submitted for the Degree of Master of Technology in Chemical Engineering Technology at The University of Johannesburg. It has not been submitted before for any degree or examination in any other University. I further declare that all sources cited or quoted are indicated and acknowledged by means of a comprehensive list of references.



.....  
John T. Kabuba

.....day of February 2009

## **Dedication**

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*I dedicate this dissertation to my Father Tshilenge Kalonji Elias who encouraged me enormously.*



## **Abstract**

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Essential oils are volatile oils, generally odorous, which occur in certain plants or specified parts of plants, and are recovered by accepted procedures, such that the nature and composition of the product is, as nearly as practicable, unchanged by such procedures (ISO, 1968). The principal uses are as: flavouring agent, medicinal and aromatherapy application. Today, the essential oils are sought-after for innumerable applications starting from markers for plant identifications to bases for semi-synthesis of highly complex molecules. The extraction of highly delicate essential oils from plants remains a crucial step in all these applications. By using steam to mediate the extraction, it is possible to maintain mild conditions and effect superior extraction.

In the current work, an integrated procedure for steam extraction followed by volatiles sampling and analysis from the leaves of the Eucalyptus tree was explored. There are two problems to overcome in the extraction from solid plant materials: that of releasing the essential oils from solid matrix and letting it diffuse out successfully in a manner that can be scaled-up to industrial volumes. Towards this end, the effect of different parameters, such as temperature, pressure and extraction time on the extraction yield was investigated and the experimental results show that all of these temperatures ( $90^{\circ}\text{C}$ ,  $97^{\circ}\text{C}$ , and  $99^{\circ}\text{C}$ ), were significant parameters affecting yield. Increase in yield was observed as pressure was increased and the use of high pressure (150 kPa) in steam extraction units permits much more rapid and complete distillation of essential oils over atmospheric pressure. The yield was calculated from the relation between the essential oil mass extracted and the raw material mass used in the extraction. The volatiles, Eucalyptus oil in vapour form released from the leaves were condensed and analyzed using Gas chromatography, and eight major components were found to be contained in this species.

A mathematical model based on diffusion of essential oil from the leaves was developed. Using a numerical method, the best diffusion coefficient was established for different operating conditions by comparing the model concentration of oil remaining in the leaves with the experimental amount of oil recovered; hence minimizing the sum of squared errors. It was found that one can not simply assume that the oil leached and recovered was the same as that originally present in the leaves. The initial mass of oil was determined by fitting the diffusion model to the data.

An Arrhenius model was used to account for the effect of temperature. The resulting expression for the diffusion coefficient as a function of temperature can now be used to model the large scale extraction of the essential oils from Eucalyptus leaves.



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## Nomenclature

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### Symbols

e Base of the natural logarithm system

$E_A$  Activation energy (kJ/mol)

Hz Hertz

$\Sigma$  Summation

g Gram

l Litre

ln Natural logarithm

D Diffusion coefficient ( $m^2/s$ )

$D_0$  Maximum diffusion coefficient ( $m^2/s$ )

C Concentration ( $mol/m^3$ )

$C_0$  Initial concentration ( $mol/m^3$ )

°C Centigrade degree

K Kelvin

Kg Kilogram

m Meter

M Mega

$M_0$  Initial mass

min Minute

Mm Molecular mass (mol/g)

x Position

t	Time
T	Temperature
$\pi$	Pi (3.14)
P	Pressure (kPa)
R	Gas constant (R= 8.314 J/mol K)
r	Correlation coefficient
$\mu$	Micro
Wt	Weight
W	Watt

## Abbreviations

CO <sub>2</sub>	Carbon Dioxide
EDA	Electro-dermal activity
EO	Essential Oils
FID	Flame Ionisation Detector
GC	Gas Chromatograph
HAZOP	Hazardous and Operability
ISO	International Standards Organization
RPM	Revolution Per Minute
SC-CO <sub>2</sub>	Supercritical Carbon Dioxide
SSE	Sum Square Errors

# **Chapter 1**

## **Introduction**

---

### **1.1 Background of essential oils, rationale and motivation**

Essential oils are volatile oils, generally odorous, which occur in certain plants or specified parts of plants, and are recovered by accepted procedures, such that the nature and composition of the product is, as nearly as practicable, unchanged by such procedures.

It specifies clearly that the nature and composition of the oil must be unchanged by the process of extraction, which is one reason why steam is an appropriate method of extraction. Furthermore, because steam distillation has been the extraction method for most essential oils, the market accepts steam distilled oil as normal oil. Oil derived by another technique might be of slightly different chemical composition and therefore might not be accepted by the market as normal oil. [ISO, 1968]

Essential oils are used widely by the pharmaceutical and cosmetic/ perfumery industries as well as in aromatherapy and alternative medicine.

Essential oils have some distinctive characteristics, which make them a very valuable commodity with many industrial uses and applications. Their aromatic value enables them to be used as flavourings in both the food and beverage industries. These oils are also widely used in both the cosmetic and pharmaceutical industries [Wonwood, 1990].

With such applications, there is a huge demand for essential oils worldwide and hence they have been traded internationally for several centuries. There is hence a need to improve the quality and quantity of the essential oils produced as they have a very competitive and profitable market worldwide [Learmonth, et al. 2002]. The chemical composition of the essential oils is important

in determining their quality and consequently price in the market [Learmonth et al. 2002]. It is therefore important to note and understand some of the parameters such as temperature, pressure, and time of extraction may affect the quality and yield of essential oil. Essential oils can be extracted using a variety of methods, although some are not commonly used today. Currently, the most popular method of extraction is steam extraction, but as technological advances are made more efficient and economical methods are being developed. These include methods such as solvent extraction, supercritical fluid extraction, cold pressing and microwave extraction. The suitability of extraction method varies from plant to plant and there is significant differences in the capital and operational costs associated. South Africa currently imports essential oils for the pharmaceutical and cosmetics industries. This project seeks to provide import substitution by enabling the development of small-scale process plants to extract essential oils from local plants using steam extraction. The technology could be transferred to interested private sector or other agencies. The government's poverty alleviation program could be well supported by the proliferation of these small-scale process plants throughout South Africa through the Technology Station Program.

## 1.2 Aims and Objectives

This work seeks to investigate steam extraction of Essential Oils and optimization of the important process parameters involved in the extraction.

Accordingly, the objectives are as follows:

1. Investigate the production of essential oils using steam extraction of fresh plant material.
2. Determine the effect of various operating parameters such as temperature, pressure and extraction time on the quality and quantity of the Essential Oils produced.

3. Setting up a model that can be used to optimize process parameters and to design steam extraction units.

In order to achieve these aims, the following methodology shall be adapted:

- (i) The quality and quantity of the extraction oil will be analysed.
- (ii) The mathematical modelling of steam distillation is necessary to design industrial plant with good operational conditions.
- (iii) Recommendations are to be made for the most optimum process parameters for steam extraction of Essential Oils.

### **1.3 Format of the study**

This research report consists of 6 chapters.

Chapter 1. Is a general introduction, which equips the reader with the necessary background, required information to place the research problem into context the aims and objectives of this study are presented.

Chapter 2. Presents an overview of the relevant literature concerning Eucalyptus and highlights the importance of these plants, also in terms of their applications. The chapter deals with the essential oil, giving a depth background into the chemical components present in essential oil, as well as the factors which influence the yield and chemical composition and methods of extraction.

Chapter 3 gives the techniques of steam extraction of essential oils and the basic scientific principles involved in this process.

Chapter 4. The overview on the materials and treatment methods used the general description of equipment and procedure; the condensation isolation and characterization of the oils extract is dealt with in chapter four.

Chapter 5. The qualitative, quantitative, and mathematical modelling results and discussions are reported in this chapter.

In the final Chapter 6, the conclusions drawn from the results are consolidated and anticipated future research emanating from this investigation is outlined.



## **Chapter 2**

### **Literature review**

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#### **2.1 History of Essential Oils**

##### **2.1.1 General history**

*Ex Oriente Lux. "The sun rises in the East".* Symbolically this old saying glorifies the East as the cradle of civilisation. In the east also began the history of essential oils; for the process of distillation the technical basis of the essential oil industry was conceived and first employed in the Orient, especially in Egypt, Persia, and India. As in many other fields of human endeavour, it was in the Occident, however, that these first attempts reached their full development. Data on the methods, objectives and results of distillation in ancient times are scarce and extremely vague. Indeed, it appears that the only essential oil of which the preparation (by a somewhat crude distillation) has been definitely established is the oil of turpentine and, if we care to mention it in connection with essential oils, camphor [Guenther, 1960].

Early in history, man exhibited a great deal of interest in the preservation of the fragrances emitted by plants, and those who were later to be called chemists occupied themselves with separating the essence from perishable plants [Guenther, 1960].

It was probably observed that heating of the plant caused the odoriferous components to evaporate, and that upon cooling and subsequent condensation, droplets united, forming a liquid consisting of two layers-water and oil. While, in such primitive experiments, the water from the plant is used to carry over the oils, additional water or steam was later introduced in extraction chamber to obtain better yields and quality [Guenther, 1960].

## 2.1.2 History of Eucalyptus Essential Oils

In this work, the emphasis is on Eucalyptus oil, one amongst the many others. This is because of its extensive availability throughout South Africa and the wide use of its essential oils. The genus names Eucalyptus is derived from the Greek words **eu**, meaning “well”, and **Kalyptos**, meaning “cover”. **Eucalyptos** refers to the well-covered mature flowers. Eucalyptus essential oils are usually extracted from Eucalyptus *Globulus* a member of the Myrtaceae family and are also known as the (Tasmania) Blue Gum [Wordiq, 2005].

Eucalyptus is a diverse genus of trees, the members of which dominate tree flora of Australia. There are almost 600 species of Eucalyptus, mostly native to Australia. They can be found in almost every part of that continent, adapted to all of Australia's climatic conditions; in fact, no other continent is so characterised by a single genus of tree as Australia is by Eucalyptus. All Eucalyptus species are evergreen, although some species have some deciduous attributes [Wordiq, 2005]. Eucalyptus has been planted and propagated in many countries of Africa, Asia and Latin America. South Africa has different species of eucalyptus across the country.

Eucalyptus leaves contain essential oils which, if emitted rise above the bush to create the characteristic distant blue haze of the Australian landscape. Eucalyptus oil is highly flammable and bush fires can travel through the oil-rich air of the tree crowns with an explosive power that can hardly be controlled.

The Eucalyptus story began in 1788 with the arrival of the first fleet and Surgeon-General John White. Within a few weeks of arriving, White recorded in his diary the presence of olfactory oil in the eucalyptus; the genus being named eucalyptus by the French botanist T, Heritier in the same year. *Eucalyptus Globulus* was discovered in Tasmania (Australia) in 1792 by Labillardiere and is commonly known as the “Blue Gum”. No Eucalyptus species has received so much attention from

botanist and chemists as this one. It has been cultivated in all parts of the world and eucalyptus oil from this eucalyptus species is the best known and most used of all Eucalyptus oils.

As a result of the collaboration with Van Meuller the essential oil industry of Australia began in 1852, when Bosisto commenced operation in small, rudely constructed still at Dandenong Creek, Victoria, using the leaves of a form of *Eucalyptus radiate*. (Known as *Eucalyptus amygdalina*) this grew profusely in the district. It is difficult to be certain which was the next species to be exploited as *Eucalyptus globulus*, *Eucalyptus oleosa* and *Eucalyptus cneorifolia* all were distilled for commercial purposes in the early 1880's.

Essential Oils are the volatile components of aromatic plants and are widely used in the pharmaceutical industry. Many farmers in Tasmania were distilling *Eucalyptus globulus* at about the same time. *Eucalyptus cinerea* was distilled in the Goulburn district of NSW between 1880 and 1900. There is no doubt that considerable interest was exhibited in the exploitation of *Eucalyptus Globulus*, which ranked only second to *Eucalyptus amygdalina* as a commercial oil-producing species. Practically all of these species, with the exception of *Eucalyptus Globules*, have been superseded by those giving larger yields of oil.

By 1900 Australia's eucalyptus oil industry was well established and able to supply the world market with substantial quantities of various types of high grade oils. By about 1950 and due to high production costs, Australia could no longer compete against Spanish and Portuguese oils and lost its leading position in the eucalyptus field.

The Eucalyptus tree can sometimes reach a height of 100 meters (30 feet), making it one of the tallest trees in the world. It uses a lot of water while growing, thus it draining land so that it sometimes has the beneficial effect of reducing the breeding area for Malaria mosquitoes aiding a healthier climate for living. It is also valuable source of hardwood and although the leaves of all species contain some Eucalyptus oil, less than 20 have enough oil of commercial value to be exploited and of these only 10 accounts for almost the entire world production. As a general rule

good timber producing eucalyptus contains very little oil and those used for their oil are of little use as timber.

All Eucalyptus oils are not the same. Each species produces an oil of different chemical composition and the constituents of oil of one species may be completely different from that of another species. However, eucalyptus oil from the same species is generally remarkably constant in its constituents and chemical composition.

Commercially important Eucalyptus oils of commerce can be grouped according to their uses into three classes, viz. medicinal, industrial and perfumery oils. Perfumery oils account for only a small fraction of the usage while medicinal and industrial oils are used in about equal proportions [Guenther, 1960].

## 2.2 The uniqueness of Essential Oils



In early work, the term “essential oils” was defined as the volatile oils obtained by the steam distillation of plants. This definition was clearly intended to make a distinction between “fatty oils” and the oils which are easily volatile.

Gradually with the advance of science came improvements in the methods of preparing the oils, and parallel with this development a better knowledge of the constituents of the oils was gained. It was found that the oils contain many classes of organic substances with varying volatility.

Although a list of all the known oil components would include a variety of chemically unrelated compounds, it is possible to classify these into four main groups of essential oils [Guenther, 1960]:

1. Terpenes, related to isoprene;
2. Straight-chain compounds, not containing any side branches;
3. Benzene derivatives;

#### **4. Miscellaneous.**

Essential oils are different from other oils by their properties:

##### **A. Essential oils are volatile:**

Essential oils are the volatile fragrant components from various indigenous and exotic plants which have been traded internationally for several centuries [Yesenofski, 2005]. All true essential oils are secondary metabolites of plant products and in some instances the oil extracted from one part of the plant is different from that extracted from other parts [Learmonth et al., 2002].

##### **B. Essential oils are aromatic**

Essential oils are highly aromatic and therefore, many of the benefits can be obtained by simply inhaling them. This can be done by breathing in the fragrance from the bottle, or they can be diffused into the room.

Essential oils, when diffused, can be the best air filtration system in the world.

They will:

- a. Purify the air by removing metallic particles and toxins from the air;
- b. Increase atmospheric oxygen;
- c. Increase ozone and negative ions in the house, which inhibits bacterial growth
- d. Destroy mold, cigarettes and animal odours;
- e. Fill the air with a fresh, herbal aromatic scent [Becker, 2005].

##### **C. Essential oils have penetrating characteristics**

The penetrating characteristic of essential oils greatly enhances their ability to be effective.

Essential oils will penetrate into the body when applied to the skin.

Essential oils rubbed into the feet will be distributed to every cell in the body in minutes.

They will even penetrate a finger or toe nail to treat fungal infection underneath. Other vegetable oils do not have this propensity to penetrate [Becker, 2005].

#### **D. Pure Essential oils have very high frequency**

The effectiveness of essential oils is sometimes also described in terms of frequency. It has been reported that the human body has an electrical frequency and that much about a person's health can be determined by frequency.

In 1992, Bruce Tainio of Tainio Technology, an independent division of Eastern State University in Cheney, Washington, built the first frequency monitor in the world.

Tainio has determined that the average frequency of the human body during the day time is 62-68 MHz. (a healthy body frequency is 62-72). When the frequency drops, the immune system is compromised. If the frequency drops to 58 MHz, cold and flu symptoms appear, at 55 MHz, diseases like Candida take hold, at 52 MHz, Epstein bar and at 42 MHz, cancer. According to Dr. Royal R. Rife, every disease has a frequency [Becker, 2005].

He found that certain frequencies can prevent the development of disease and that others would destroy disease. Substances with higher frequency will destroy diseases of a lower frequency. The study of frequencies raises important questions, concerning the frequencies of substances we eat breath and absorb. Many pollutants lower healthy frequency. Processed canned food has a frequency of zero. Fresh produce has up to 27 MHz. Essential oil start at 52 MHz and go as high as 320 MHz, which is the frequency of Rose oil. Clinical research shows that essential oils have the highest frequency of any natural substance known to man, creating an environment in which disease, bacteria, virus, fungus, etc., cannot live [Becker, 2005].

### **2.3 Uses of Essential oils**

According to ancient Egyptian hieroglyphics and Chinese manuscripts, priests and physicians were using essential oils thousands of years before Christ to heal the sick.

These are the oldest form of medicine and cosmetic known to man and were considered more valuable than gold. There are 188 references to oils in the Bible.

The wise men brought the Christ child gold, frankincense and myrrh. Current clinical research shows that frankincense oil contains a high concentration of immune stimulating properties. In Exodus, God gives Moses the formula for “holy anointing oil” (Exodus 30:22-25). Some of the precious oils which have been used since antiquity for anointing and healing the sick are frankincense, myrrh, galbanum, hyssop, cassia, cinnamon and spikenard. Science is only now beginning to investigate the incredible healing substances found in essential oils [Becker, 2005].

Essential oils have a wide range of uses:

- ❖ As an important sources of natural flavours and are used extensively in the agric-food sector.
- ❖ As fragrances and are used in perfumed, beauty products, deodorants, soap and detergents.
- ❖ As components in pharmaceuticals, antiseptic, and aromatherapy products.

Among the major uses of essential oils are:



### **2.3.1 Fragrant uses**

Currently fragrant of essential oils are mainly used by the cosmetic industry. Smell has quite an effect on the human body and its nervous responses.

The limbic system in the human body is represented by the hippocampus, formic, cingulated gyrus, thalamus, mamillary bodies, amygdale and olfactory bulb. And although this might sound far too technical to be interesting, it is fascinating how the sense of smell actually happens and the effect it has on us all. Before the term “limbic system” was used, the system was referred to as the *rhinencephalon* or “smell brain” and is considered as one of the oldest systems in the human body.

When a smell enters the nasal cavity, it meets up with over 50,000,000 receptor neurons which are located in the upper part of the nose and nasal septum, and the receptors are specialized in such a way that certain react to certain smells.

These receptors then convert the presence of a smell into a message which is sent to the olfactory bulb (which is seen as part of the forebrain and its main sensory input) where the processing of the smell is started and then passed onto other areas of the brain which control emotions, behaviour as well as basic thought processes. Some distant memories, for instance from childhood, can only be recalled by smell, and this fact underscores the importance and significance of smell for our mental life and well-being.

### **Brainwaves**

When measuring brainwaves by means of an electroencephalogram (EEG) it has been noted that there was an increase of alpha waves when smelling lavender oil, which is assumed to be a relaxant fragrance. These test results did however vary when test subjects were in different states of arousal or relaxation. When using alerting odours, such as jasmine, an increase in brain activity was noted. On the other hand, certain odours such as nutmeg, mace extract and valerian oil reduced systolic blood pressure and stress. Interestingly an odour need not be very strong to have an effect on the body. With electro-dermal activity (EDA) testing (where the electrical current between two points of the skin is measured) it was also found that slower currents occurred with relaxing odours such as bergamot and lavender were inhaled.

### **Hedonics**

It must however be kept in mind that hedonics play a large part in interpreting the results of such tests. Hedonics is the personal degree of pleasantness that a person would place on a specific odour or smell.

The limbic system is more than just a part of our smelling mechanism: it is an integral part of man in the wider context. Since it has a direct and indirect influence on so many of our body systems, including the regulation of the endocrine and visceral effect or mechanisms and the resultant patterns of behaviour and motivation, it therefore implies that our sense of smell is more than simply a coping mechanism, but fulfils its own regulatory work as well.

Although much scientific research has been done on how odours, and in particular the fragrances of essential oils, influence the physiology of the body, much is still unexplained and calls for further investigation.

### **2.3.2 Medicinal uses**



The principal active ingredient in eucalyptus oils is “eucalyptol” which has strong germicidal and disinfectant properties.

Medicinal eucalyptus oil produced from *Eucalyptus polybractea* is widely used for the relief of cold and influenza symptoms. It is a unique natural product having antiseptic properties and the power to clear the nasal passages and bronchial tubes making it easier to breathe. A popular new use is to vaporise it in saunas. It is an excellent rub for muscular aches and pains and it has been widely used for many years by sportsmen to help keep muscles trim and supple. A use which is gaining widespread acceptance is the practice of adding eucalyptus oil to the laundry wash for cleaning and freshening clothes, which utilises its cleaning, deodorizing and antiseptic properties.

Medicinal eucalyptus oils and eucalyptol are extensively used as a raw material and active ingredient of cough lozenges, inhalation sprays and drops, gargles, mouth washes, toothpastes, embrocating balms and ointments, liniments and soaps. Eucalyptus oils are also used as antiseptic and germicidal disinfectants because of its pleasant odour and its effectiveness in killing bacteria. It is an excellent solvent which makes it an ideal spot and stain remover [Peter, 2004].

Eucalyptus has a cooling and deodorizing effect on the body, helping with fevers, migraine and malaria. For the respiratory tract, it helps with coughs, asthma, throat infections, sinusitis and catarrhal conditions. It soothes inflammation and eases mucus, clearing the head from the stuffiness of colds and hay fever. Eucalyptus oil is useful as warming oil when used for muscular aches and pains, rheumatoid arthritis, sprains and poor circulation.

In skin care it can be used for burns, blisters, helps, cuts, wounds, skin infections and insect bites. Eucalyptus oil can boost the immune system, and is helpful especially in cases of chickenpox, colds, flu and measles. The oil is also effective against bacteria-especially staphylococci. In vapour therapy, eucalyptus oil is used for frequent sneezing, hay fever, flu, respiratory problems and as insect repellent. Eucalyptus oil is also used in blended massage oil, or diluted in the bath to assist with: arthritis, asthma, bronchitis, mucous congestion, colds, headaches, rheumatism, sinusitis, catarrh, fatigue and muscular aches and pains [Crowley, 2001].

## **2.4 Methods of production**

Essential oils can be extracted using a variety of methods, although some are not commonly used today. The specific extraction method employed is dependant upon the plant material to be distilled

and the desired end-product. The essential oils from aromatic plants are for the most part volatile and thus, lend themselves to several methods of extraction such as steam distillation, solvent extraction, supercritical fluid extraction, etc.

### **2.4.1 Steam Distillation**

The vast majority of essential oils are produced by steam distillation. There are, however, different processes that are used. In all of them, water is heated to produce steam, which is used to extract the most volatile aromatic chemicals. The steam is then cooled (in a condenser) and the resulting distillate is collected. The essential oils will normally float on top of the hydrosol (the distilled water component) and may be separated off. Steam distillation is the most commonly used method for extracting essential oils. Many traditional distillers favour this method for distilling most oils as they claim that none of the newer methods produce better quality oils [Boucard et al., 2005].

Steam distillation, as described by Boucard et al. [2005], is done in a still in which fresh or sometimes dried plant material is placed in a chamber of the still. Pressurized steam, generated in a separate chamber, is then circulated through the plant material. The heat of the steam forces open the tiny intercellular pockets in which the essential oils are contained releasing the oils. During steam distillation, the temperature of the steam must be moderated so that it is high enough to open the oil pouches without destroying the plants, fracturing or burning the essential oils as has been recommended in the literature [Sheridan, 2000]. Some or most essential oils have been found to be heat sensitive and hence thermo degradable.

As the tiny droplets of essential oils are released, they evaporate and mingle with the steam, travelling through a pipe into a condenser. The steam and oil vapour are then condensed to a liquid mixture.

As the oil-water mixture has been found to be nearly immiscible at a temperature lower than about 65°C in previous work done by Sheridan [2005], and the mixture can be separated using various gravity related techniques. Due to the immiscibility of the oil and water at low temperature, the essential oil can be separated from the water by either decanting off the water or skimming of the oil from the top as the oil is less dense than water at these conditions. The density of some essential oils such as lavender oil has been reported to average 0.89g /l, as opposed to 1g /l [Ndou, 1986] for water at room temperature and atmospheric pressure conditions. The water obtained as a by product of distillation is referred to as floral water or distillate and retains many of the therapeutic properties of the plant. For this very reason, floral waters are valuable in skin care for making facial mists and toners and are also preferred to essential oils when treating a sensitive individual or child or when a more diluted treatment is required [Sheridan, 2000].

#### **2.4.2 Solvent Extraction**

The purpose of distillation is to separate a mixture of several ingredients by taking advantage of their different volatility, or to separate volatile ingredients of a raw material from its non-volatile parts. If the final product is too sensitive to heat or humidity, solvent extraction could be used.

Solvent extraction is adapted in producing essential oils generated by some flowers (Rose, Violetta, and Geranium), gums and resins. The raw material is placed in a glass, vessel and soaked with a suitable solvent (petroleum, ether or benzene). After the extraction, the solids are separated from the liquid mixture. The latter is heated so that the more volatile essential oils can be

evaporated to be subsequently condensed. Alternatively if the solvent is more volatile, such as ethanol, it could then be vaporised leaving behind the essential oils [Ndou, 1986].

As solvent extraction uses very little heat, it is found to be advantageous in producing essential oils with whole fragrances that would otherwise be destroyed or altered during steam distillation. Therefore this extraction technique can be used to extract essential oils from very delicate plants to produce higher amounts of essential oils at lower costs [Ndou, 1986]. There are, however, some disadvantages associated with the solvent extraction technique. Solvent residues often contaminate the product causing side effects which make the use of essential oil undesirable for skin applications but could still be fine for fragrances or perfumes [Ndou, 1986]. Therefore with solvent extraction effective separation of the extracted oil from the solvent is necessary to remove any solvent which may contaminate the essential oils.

This process also sometimes yields an aromatic resinous product known as oleoresin which is more concentrated than essential oils with an even wider application in the food and other industries, as discussed by Heath [1981].

#### **2.4.3 Supercritical fluid extraction**

Supercritical fluid extraction (SFE) of essential oils is a modern technique, currently being applied in the process industry, which competes with conventional processes such as steam distillation and organic liquid (solvent) extraction. It has been widely accepted by many investigators that SFE provides a rapid and quantitative method for extracting essential oils from aromatic plants that compares favourably with steam distillation [Kerrola, 1995]. A single-component fluid is said to be supercritical when its temperature and pressure both exceed their critical values, without being far

from the critical state [Gaspar et al., 2002]. At these elevated conditions the properties of the fluid has both liquid and gas properties.

All materials have a critical point, but for some materials however this state is more easily reached than others. Carbon dioxide is the most commonly used fluid in extracting essential oils and its application and technique have been extensively researched. Supercritical extraction by carbon dioxide is isolation and separation process taking advantage of the fact that above critical conditions of 31.1°C and 78.8 bar, carbon dioxide cannot be liquefied by any further increases in pressure. Whilst the carbon dioxide is in this supercritical state, the dense gas gains a considerable solvent power, dissolving the primary target such as the essential oil in the plant material. The pressure is then dropped in the separator, causing the carbon dioxide to lose its solvating power and hence releasing the extracted oil drops, leaving behind a high purity essential oil extract. This high purity of these products has recently attracted the attention of the pharmaceutical industry, due to the heavily regulated trade demands for lower quantities of solvent in the final product. This area of research is currently being pursued and analysed by various researchers such as [Gaspar et al., 2002], in their work for various pharmaceutical companies worldwide, in developing new production techniques based on supercritical carbon dioxide.

Using carbon dioxide is especially useful as it is cheap, clean and intrinsically safe with no harmful residues like solvents in the extracts that would be found for solvent extraction as discussed before. The operating temperatures are relatively low, which enables thermally labile compounds to be extracted. This method of extraction has also been reported to ecologically harmless in literature [Gaspar et al., 2002] as no harmful residues of toxic solvents result from it.

Supercritical fluid extraction by carbon dioxide has therefore been described by [Simon, 1990] as a new method; potentially commercially viable but is less common and beyond the financial means of most small scale processors.

#### **2.4.4 Cold pressed expression method**

Another method of extraction essential oils as described in the literature [Stahle et al., 1988] is cold pressed expression, or scarification. It has been reported to be used mainly to obtain citrus fruit oils such as bergamot-, grapefruit-, lemon-, lime-, mandarin-, orange-, and tangerine oils [Wu et al., 2001].

In this process, the fruit is rolled over a through with sharp projections that penetrate their peels thereby piercing the tiny pouches containing the essential oil. The fruit is then pressed to squeeze out the juice from the pulp thereby releasing the essential oils from the pouches. The essential oils rise to the surface of the juice and are separated by centrifugation.

As discussed in the literature [Stahl et al., 1988], cold pressing is more competitive for specific raw material than methods such as supercritical fluid extraction as it is extremely fast, cheap and does not pollute the extracts, although it does not provide a way of selectively extracting essential oils.

#### **2.4.5 Effleurage Method of Extraction**

Some flowers, such as jasmine or tuberose, have such low contents of essential oils and are so delicate that heating them would destroy their blossoms before releasing the essential oils. In such cases, an expensive and lengthy process called effleurage is sometimes used to extract the essential oils. As described in the literature [Stahl et al., 1988], flower petals are placed on trays of odourless vegetable or animal fat, which absorb the essential oils from them. At the end of every day or even after a few hours, when the vegetable or fat has removed as much of the essential oil as possible, the depleted petals are removed and replaced with fresh ones. This procedure is repeated until the fat or oil becomes saturated with the essential oil. Adding alcohol to this effleurage mixture

separates the essential oils. This method employs a similar operating principle and technique to what was discussed for solvent extraction.

#### **2.4.6 Microwave Extraction**

Microwave energy is a superior alternative to several thermal applications owing to its efficient volumetric heat production. The volumetric heating or heating of the bulk as opposed to transferring heat from the surface, inwards, is more efficient, uniform and less prone to overkill or supererogation. Controllability is by far the greatest advantage of microwaves over conventional thermal technologies. In processing applications, the ability to instantaneously shut the heat source makes enormous difference to the product quality and hence the production economics. The raw material is heated directly by microwaves and this brings about quality consistency and minimizes the impact on the environment as apposed to using fossil fuels or less efficient, indirect electrical heating systems. Specifically in the essential oil extraction, microwave mediated processes are highly desirable due to their small equipment size (portability) and controllability through mild increments of heating. However, so far the microwave technology has found application in very few industrial bio-processing installations due to the lack of available data on microwave interaction with heterogeneous natural raw materials.

The sensing and close control of microwave process is a challenging science and there seems to be insufficient literature in this regard.

## **2.5 Important Physical and Chemical properties of Essential Oils**

The chemical properties of essential oils depend on the natural factors such as type of species, the geographical origin and location of the plant, time of harvesting, plant parts from which the oils are extracted, etc[Dey, 1996].

Essential oils components and percentage are different from oil to oil even for the same botanic plant due to:

**a) Weather and planting time**

Most of herbs are planted but small amounts could also be wild grown or collected plants. By means of an example with spearmint, the oil percentage from a summer crop is double that from a winter crop. The oil percentage from a given summer could be different from a previous summer even from the same field. The component analysis of the oil could also be different from one season to another.



**b) Soil elements**

The B-phellanderene percentage increases in marjoram oil with the higher levels of molybdenum manganese, copper, calcium, zinc or iron in the soil.

**c) Irrigation**

The highest yield of plant material results from increasing the leaf area. For example, this will happen if a basil field is irrigated every 4 days. The essential oil is highest at medium levels of soil moisture.

**d) Time of harvest**

The peppermint oil yield increases as the herb approaches maturity in the full bloom stage.

### e) Length of distillation operation

To specify time for distillation operation you must consider whether the herbs (species) are fresh, faded, or dry. It would take additional time for distillation if the herb is faded than if it is fresh.

Leaves take less time than seeds because leaves are thinner than seeds and cells are more concentrated in leaves than seeds [Guenther, 1960].

## 2.5.1 Physical properties

### 2.5.1.1 Specific gravity

Specific gravity is an important criterion of the quality and purity of an essential oil. Values for essential oils vary between the limits of 0.696 and 1.188 at 15°C, in general, the specific gravity is less than 1.000 [Guenther, 1960]. Hence essential oil can be collected over (floating on) water.

### 2.5.1.2 Optical rotation

Most essential oils when placed in a beam of polarized light possess the property of rotating the plane of polarization to the right (dextrorotatory), or to the left (laevorotatory). The degree of rotation and the direction are important indicators of purity.

### 2.5.1.3 Refractive index

When a ray of light passes from a less dense to a more dense medium, it is bent or “refracted” toward the normal. If  $e$  represents the angle of refraction and  $i$  the angle of incidence, according to the law of refraction,

$$\frac{\sin i}{\sin e} = \frac{N}{n}$$

Where  $n$  is the index of refraction of the less dense, and  $N$ , the index of refraction of the denser medium.

Refractometers offer a rapid and convenient method for the determination of this physical constant.

#### **2.5.1.4 Molecular refraction**

The index of refraction of a liquid varies with temperature and the wave length of the light. In order to compare the refractivities of different liquids, the use of molecular refractivity (molecular refraction) is necessary.

#### **2.5.1.5 Solubility**

- **Solubility in Alcohol**

Most essential oils are only slightly soluble in water and are miscible with absolute alcohol.

The solubility of oil may change with age.

- **Solubility in water**

Most of essential oils of commercial interest are steam volatile, reasonably stable to action of heat and practically insoluble in water and hence suitable for processing by steam distillation.

#### **2.5.1.8. Boiling range**

In the case of isolates and synthetics, the boiling range is an important criterion of purity.

### **2.5.1.9. Evaporation residue**

An important criterion of purity is the evaporation residue; i.e., the percentage of the oil which is not volatile at 100°C.

It is important to study the odour of oil as it volatilizes during the heating.

### **2.5.1.10. Flash point**

The flash point may prove useful in the valuation of an essential oil. The flash point has value as an indication of adulteration: additions of adulterants such as alcohol and low boiling mineral spirits will greatly lower the flash point.

## **2.5.2 Chemical properties**

In general, essential oils consist of chemical compounds that have hydrogen, carbon, and oxygen as their building blocks. These can be subdivided into two groups: the hydrocarbons, which are made up almost exclusively of *terpenes* (*monoterpenes*, *sesquiterpenes*, and *diterpenes*); and the oxygenated compounds, mainly esters, aldehydes, ketones, alcohols, phenols, and oxides; acids, lactones, sulphur and nitrogen compounds are sometimes also present.

**2.5.2.1 Aldehydes:** citral, citronellal, and neural are important aldehydes found notably in lemon-scented oils such as Melissa, lemongrass, lemon verbena, citronella, etc.

Aldehydes in general have a sedative effect; *citral* has antiseptic properties.

**2.5.2.2 Phenols:** These tend to have a bactericidal and strongly stimulating effect, but can be skin irritants. Common phenols include *eugenol* (found in clove and West India bay), *thymol* (found in thyme), *carvacrol* (found in oregano and savoury).

**2.5.2.3 Terpenes:** common terpene hydrocarbons include limonene (antiviral, found in 90 per cent of citrus oils) and pinene (antiseptic, found in high proportions in pine and turpentine oils). Sesquiterpenes have outstanding anti- inflammatory and bactericidal properties.

**2.5.2.4 Ketones:** some of the most common toxic constituents are ketones, such as thujone found in *mugwort*, tansy, sage and wormwood; and *pulegone* found in pennyroyal and *buchu*. Non-toxic ketones include jasmine (in Jasmine) and *fenchone* (in fennel oil).

**2.5.2.5 Oxides:** by far the most important oxide is cineol (or eucalyptol). It has an expectorant effect, and is well known as the principal constituent of eucalyptus oil.

It is also found in a wide range of other oils, especially those of a *camphoraceous* nature such as rosemary, bay laurel, tea tree, and cajuput.

**2.5.2.6 Esters:** probably the most widespread group found in essential oils, which includes linalyl acetate (found in bergamot, clary sage, and lavender) and geranyl acetate (found in sweet marjoram). They are characteristically fungicidal and sedative, often having a fruity aroma.

**2.5.2.7 Alcohols:** these compounds have good antiseptic and antiviral properties with an uplifting quality; they are also generally non-toxic. Among the most common terpene alcohols are *linalool* (in rosewood, *linaloe*, and lavender), *citronellol* (in rose, lemon, eucalyptus and geranium) and *geraniol* (found in palmarosa); also *borneol*, *methol*, *terpineol*, *nerol*, *farnesol*, *vetiverol*, benzyl alcohol, and *cedrol* [Lawless, 2002].

## **2.6 Factors affecting the yield and quality of the Essential Oils**

The yield and quality of essential oils have been known to vary due to a number of factors.

### **❖ Mode of Distillation**

Technique for the distillation should be chosen on basis of oil boiling point and nature of herb as the heat content and temperature of steam can alter the distillation characteristics.

### **❖ Proper design of equipment's**

Improper designing of tank, condenser, or separators can lead to loss of oils and high capital investments.

### **❖ Material of Construction of equipment's**

Essential oils which are corrosive in nature should be preferably distilled in stills made of resistant materials like aluminium, copper or stainless steel.

### **❖ Condition of raw material**

Condition of raw material is important because some materials like roots and seeds will not yield essential oil easily if distilled in their natural state. These materials have to be crushed, powdered or soaked in water to expose their oil cells.

### **❖ Filling of raw material / steam distribution**

Improper loading of the herb may result in steam channelling causing incomplete distillation.

❖ **Operating parameters like steam injection rate inlet pressure / condensate temperature**

Proper control of injection rates and pressure in boiler operated units is necessary to optimize the temperature of extraction and for maximal yield temperature of condensate should not be too high as this can result in oil loss due to evaporation.

❖ **Time given for distillation**

Different constituents of the essential oil get distilled in the order of their boiling points. Thus the highest boiling fractions will be last to come over when, generally, very little oil is distilling. If the distillation is terminated too soon, the high boiling constituents will be lost.

❖ **Pre condition of tank / equipments**

Tanks should be well steamed for multiple crop distillation. Tank / equipments should not be rusted for quality oil [Dey, 1998].

## **Chapter 3**

### **Principles of Steam Extraction of Essential Oils**

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#### **3.1 Preamble**

Most essential oils are obtained from plant material by a process known as steam distillation. Steam distillation is based on the assumption that essential oils are volatile in steam and generally insoluble in water.

The fundamental nature of steam distillation is that it enables a compound or mixture of compounds to be distilled (and subsequently recovered). In the presence of steam, these substances are volatilized at a temperature close to 100°C at atmospheric pressure.

The mixture of hot vapours will, if allowed to pass through a cooling system, condense to form a liquid in which the oil and water comprise two distinct layers.

Most (but not all) essential oils are lighter than water and form the top layer. The steam that is used for the distillation is generated either within the vessel that contains the plant material (by boiling water contained at the base) or by an external boiler.

The use of steam generated within the vessel requires that the plant material be supported above the boiling water by a grid. The water is heated either directly using a burning fuel or by heat exchanger coils. The simplicity of the method makes it suitable for small-scale industrial production.

If steam is generated by an external boiler, it is introduced into the base of the vessel via an open coil, jets or similar device(s). The advantages of this type of generation are that it is relatively rapid and allows greater control by the operator.

The vessel can be emptied and recharged quickly. With the immediate reintroduction of steam there is no unnecessary delay in the commencement of the distillation process.

## **3.2 Components of the Extraction Plant**

Steam distillation process for the extraction of essential oils from plant materials consists of four basic parts:

### **3.2.1 Steam Generator or Boiler**

The dictionary defines steam as “the gas or vapour into which liquid water is changed by boiling, especially when used under pressure as a source of energy”.

The primary objective of a boiler operation is to provide a continuous supply of steam at whatever pressure and temperature are suitable for the end use.

The secondary objective is to provide such steam at the lowest possible cost.

These objectives translate into “operational efficiency”.

There are many boilers available for steam generation. Some are high pressure boilers and other medium to low pressure boilers. Most process steam in industries falls under medium / low pressure. For the purpose of essential oil production, low pressure steam is generally used. The capacity of the boiler will depend on the size of distillation units [James et al., 1926].

### **3.2.2 Extraction Chamber**

The extraction chamber is simply a fixed bed where the hot steam helps to release the aromatic molecules from the plant material since the steam forces open the pockets in which the oils are kept in the plant material. The molecules of these volatile oils then escape from the plant material and evaporate into the steam. The temperature of the steam needs to be carefully controlled at a temperature just high enough to force the plant material to let go of the essential oil, yet not too high as to burn the plant material or decompose the essential oil. The steam, which then contains the essential oil, is passed through a cooling system to condense the steam, which forms a liquid from which the essential oil and water are then separated. The steam is produced at slightly greater pressure than the atmosphere and therefore boils at above 100°C, which facilitates the removal of the essential oil from the plant material at a faster rate and in so doing prevents damage to the oil [James et al., 1926].



### **3.2.3 Heat Exchanger**

Heat exchanger is a device that provides the flow of thermal energy between two or more fluids at different temperatures. The vapours from extraction chamber are cooled by passing them down a tube that is immersed in a flow of water.

The condensate runs by gravity into a container in which the oil and water separate due to their differing specific gravities: the “Separator”.

Heat exchangers are used in a wide variety of applications. These include power production; process, chemical and food industries; electronics; environmental engineering; waste heat recovery; manufacturing industry; and air-conditioning, refrigeration, and space applications. Heat exchangers may be classified according to the following main criteria:

1. Recuperators / regenerators
2. Transfer processes: direct contact and indirect contact
3. Geometry of construction: tubes, plate and extended surfaces
4. Heat transfer mechanisms: simple-phase and two-phase
5. Flow arrangement: parallel-, counter-, and cross flows

The major construction types are tubular, plate and extended surface heat exchangers.

Tubular heat exchangers are built of cylindrical tubes. One fluid flows inside the tubes and the other flows on the outside of the tubes. Tube diameter, the number of tube length, the pitch of the tubes, and the tube arrangement can be changed. Therefore, there is considerable flexibility in their design.

Tubular heat exchangers can be further classified as follows:

1. Double-pipe heat exchangers
2. Shell-and-tube heat exchangers
3. Spiral tube heat exchangers

Shell- and tube heat exchangers are built of tubes mounted in large cylindrical shells with the tube axis parallel to that of the shell. They are widely used as oil coolers, power plant, steam generators in nuclear power plants, in process applications, and in chemical industry. One fluid stream flows through the tubes while the other flows on the shell side, across or along the tubes. In a baffled shell-and-tube heat exchanger, the shell-side stream flows across between pairs of baffles and then flows parallel to the tubes as it flows from one baffle compartment to the next. There are wide differences between shell-and-tube heat exchangers depending on the application. The main design

objective here is to accommodate thermal expansion, to furnish ease of cleaning, or to provide the least expensive construction if other features are of no importance. In a shell-and-tube heat exchanger with fixed tube sheets, the shell is welded to the tube sheets and there is no access to the outside of the tube bundle for cleaning. This low-cost option has only limited thermal expansion, which can be somewhat increased by expansion bellows. Cleaning of the tube is easy. A number of shell-side and tube-side flow arrangements are used in shell-and-tube heat exchangers depending on heat duty, pressure drop, pressure level, fouling, manufacturing techniques, cost, corrosion control, and cleaning problems. The baffles are used in shell-and-tube heat exchangers to promote a better heat transfer coefficient on the shell-side and to support the tubes [James et al., 1926].

### **3.2.4 Separator**

The mixture of condenser oil and water runs into the separator where the lighter insoluble oil floats on the surface and accumulates slowly and from where it is drawn off periodically. This is the Essential oil.

The water, or rather water containing the more water soluble constituents extracted by the steam, known as the hydrolat or “floral water” is drawn off continuously from the bottom of the separator. Or, in separating the water from the oil, the water layer was carefully run out from the bottom of the decanter by opening the tap until its meniscus is just at the calibration mark. The contents that remained inside the decanter which were now the oil layer and the water between the tap bridge and the bottom of the calibration mark were weighed on the electronic analytical balance. The isolation of the water from the essential oil has to be done as carefully and accurately as possible in order to minimize the effect of human error due to any inaccuracies in levelling the water and oil layer to coincide with the level calibration mark [James et al., 1926].

### 3.3 Basic scientific principles involved in the process

In steam extraction to convert any liquid into a vapour we have to apply energy in form of heat called the latent heat of vaporization. A liquid always boils at the temperature at which its vapour pressure equals the atmospheric / surrounding pressure. For two immiscible liquids the total vapour pressure of the mixture is always equal to the sum of their partial pressures. The composition of the gas mixture will be determined by the concentration of the individual liquid components and their respective partial pressures. As known, the boiling point of most essential oil components exceeds that of water and generally lies 150-300°C.

Consider a sample of an essential oil having a component 'A' with a boiling point of 1900°C and water having a boiling point of 100°C; when these two immiscible liquids are brought together, and once their vapours have reached saturation, the temperature will immediately drop to 99.5°C, which is the temperature at which the sum of the two vapour pressure equals 760mm Hg

$$P_{Total} = P_A + P_{Water}$$

3.1

Therefore in a mixture of water and essential oil the saturated vapour above the mixture is:

$$\frac{Wt \text{ of } 'A'}{Wt \text{ of water}} = \frac{\text{vapour pressure of } 'A'}{\text{vapour pressure of water}} \times \frac{\text{Mol. Wt of } 'A'}{\text{Mol. Wt of water}} \quad 3.2$$

Thus any essential oil having high boiling point range can be evaporated with steam in a ratio such that their combined vapour pressures will be equal to the atmospheric pressure and can be isolated from the plants by the wet distillation process [Guenther, 1960].

## **3.4 Techniques of Extraction**

### **3.4.1 Water / Hydro Distillation**

Water/ Hydro Distillation is one of the simplest, oldest and primitive processes known to man for obtaining essential oils from plants and is mostly used by small scale producers of essential oils. In Water / Hydro Distillation the plant material is almost entirely covered with water as suspension in the still which is placed on a furnace. Water is made to boil and essential oil is carried over to the condenser along with the steam. This procedure is useful for distillation of powders of spices and comminuted herbs etc.

#### **Disadvantages of the Hydro Distillation**

- The process is slow and the distillation time is much longer thereby consuming more firewood / fuel making process uneconomical.
- Variable rate of distillation due to difficult control of heating.
- Extraction of the herb is not always complete.
- As the bottom walls of the still comes in direct contact with the fire from furnace there is a possibility of adjacent plant material getting charred and thus imparting an objectionable odour to the essential oil.
- Prolonged action of hot water can cause hydrolysis of some constituents of the essential oils such as esters, which reacts with the water at high temperatures to form acids and alcohols.
- Not suitable for large capacity / commercial scale distillations and not suitable for high boiling hardy roots / woody plant materials [Guether, 1960].

### **3.4.2 Steam and Water distillation**

To overcome the drawbacks of the water/ hydro distillation, the technique was modified and wet steam distillation was developed:

The plant material is supported on a cage / perforated grid below which water is boiled. Direct contact of plant material with hot furnace bottom is thus avoided. The water below the grid is heated by open fire which produces saturated and wet steam which rises through the plant material vaporizing the contained essential oil.

#### **Advantages:**

- High oil & reproducible yields, faster, lesser fuel.
- The field distillation / portable / directly fired type units are based on this technique.
- Due their very simple construction, low cost and easy operation field distillation units are extremely popular with essential oil producers in developing countries.
- Such field units generally have capacities to hold 100 kg to 2000 kg plant material.

#### **Disadvantages:**

- Unsafe, time consuming due to low pressure steam, poor quality oil.
- Improper condensation, oil separation incomplete, less recovery.
- Poor material of construction and excessive pollution [Guether, 1960].

### **3.4.3 Hydro Diffusion**

The process uses principle of osmotic pressure to diffuse oil from the oil glands. The system is connected and low pressure steam from a boiler is passed into the plant material from a boiler from the top. Pressure is applied to the plant material to force oil through a membrane. The condenser,

generally of tubular construction which is directly under the basket within the extraction chamber, is used for cooling.

The oil and water are collected below the condenser in a typical oil separator. It would appear that hydro diffusion is an efficient process to use as it gives good yield of the oil. But the co-extraction of other non volatiles and polar components with the essential oils complicates the process.

### **3.4.4 Direct steam distillation**

Also referred sometimes as dry steam distillation, plant material is supported on the grid and saturated steam from an outside source i.e. boiler is injected through a steam coil. Steam in a field distillation unit is at atmospheric pressure and hence its temperature is limited to 100°C. But steam in a modern pressure boiler operating at say 40 pounds per square inch (psi) pressure will have a temperature correspondingly higher. The use of high pressure steam in modern steam distillation units permits much more rapid and complete distillation of essential oils.

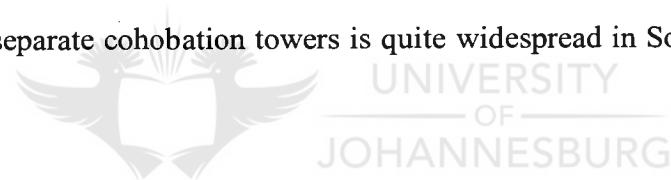
Fuel costs are generally lower in modern steam distillation units due to the higher thermal efficiency at which most of the boilers operate.

#### **Features:**

- Steam control, no thermal decomposition and suitable for large scale and multiple units.
- Capital cost is quite high so that only bigger producers can afford to own such units.
- Still capacities range from 1ton to 3 ton plant material per batch.
- Material of construction could be mild steel, or stainless steel depending upon the corrosive nature of the essential oil [Guenther, 1960].

### **3.4.5 Distillation with cohobation**

Most essential oils have finite solubility in water but in certain oils like the oil of rose the solubility is quite high. In such a situation the loss of oil taking place with the outgoing waters of distillation can become alarmingly high. This problem is solved by returning the condensed water from the separator back to the extraction chamber. It cannot be done with steam distillation as the water level in the extraction chamber will keep on building up due to continuous steam injection. Instead the distillation is carried out in the mode of water and steam distillation. Condenser is moved above the distillation still so that condensed water from separator can flow by means of gravity to the extraction chamber. By limiting the total quantity of water in this closed cycle operation, it is possible to obtain increased yields of essential oils which are more water soluble. Cohobation of distillation waters in separate cohobation towers is quite widespread in Soviet Union and Bulgaria [Guenther, 1960].



## **Chapter 4**

### **Experiments**

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#### **4.1 Introduction**

According to what has been found and discussed in literature, a number of factors individually play a significant role in the quantity and quality of essential oils extracted from plant materials. These factors include the type of extraction method and operating conditions of the processing methods, as has been mentioned and discussed. It is therefore quite evident that for a given situation when all factors come into play, their individual effect on the quantity and quality of the extracted oil cannot be quantified easily. To measure the individual effect of any of the contributing factors would require keeping all the others constant as practically possible while varying the factor being studied.

This chapter presents the experimental methods and techniques used to investigate the extraction on the yield and quality of Eucalyptus essential oils using steam extraction.

#### **4.2 Materials and methods**

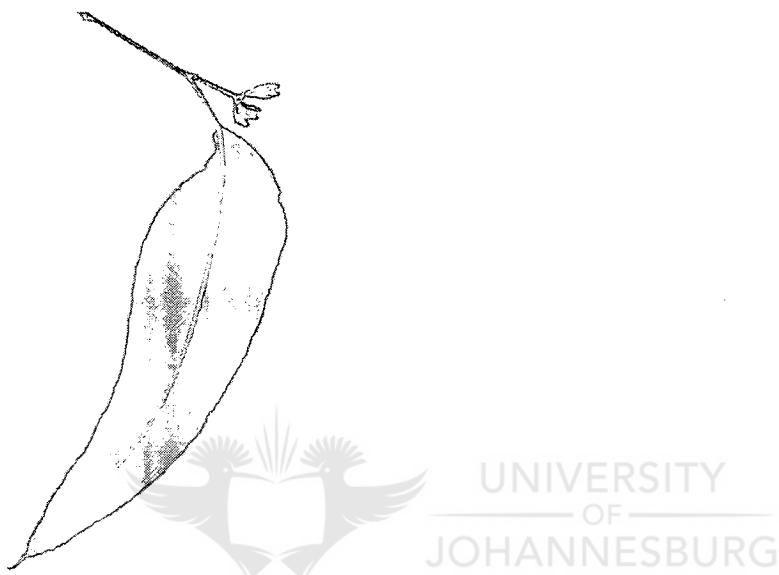
##### **4.2.1 Collection of the plant material**

A bulk sample of eucalyptus leaf materials was collected from a single location in a reserved garden in the Region 9 area of Johannesburg on the 5<sup>th</sup> day of October 2007.

The fresh plant material was taken to the laboratory of the Chemistry Department on the Arcadia Campus of the Tshwane University of Technology in Pretoria, where the leaves were picked off

the stalks and mixed thoroughly to form a homogeneous and representative stock pile sample. The mixing was to ensure that the sample had an even distribution of oil quality and yield from which samples to be treated would be taken.

#### 4.2.2 Treatment of the plant material



**Figure 4.1: Eucalyptus leaves**

Firstly, a sample with a mass of approximately 20 kg was prepared by loading on an electronic balance. The material was spread in a controlled temperature environment in a 20-30cm thick layer and left to air dry. An overnight drying period was implemented because it is reported to be the required in previous work for this process [Kasumba, 2003]. On completion of the drying process, the leaves were re-weighed using the analytical balance to determine any potential mass loss as a result of the evaporation of both moisture and volatile oil components due to the drying effect.

Secondly, the plant material was packed into the extraction chamber so that distillation could commence. Proper charging is very important; otherwise the steam channels through the plant material and low yield results.

For each load, fresh plant material (20kg) was placed into the extraction chamber. The first load was conducted to set-up and establishes the procedure and determines processing parameters. Two tests were conducted for each of three operating temperatures as part of the main study.

## 4.3 General description of equipment and procedure

### 4.3.1 Experimental and equipment description

The experimental equipment of steam extraction of essential oils is illustrated schematically in Figure 4.2 below.

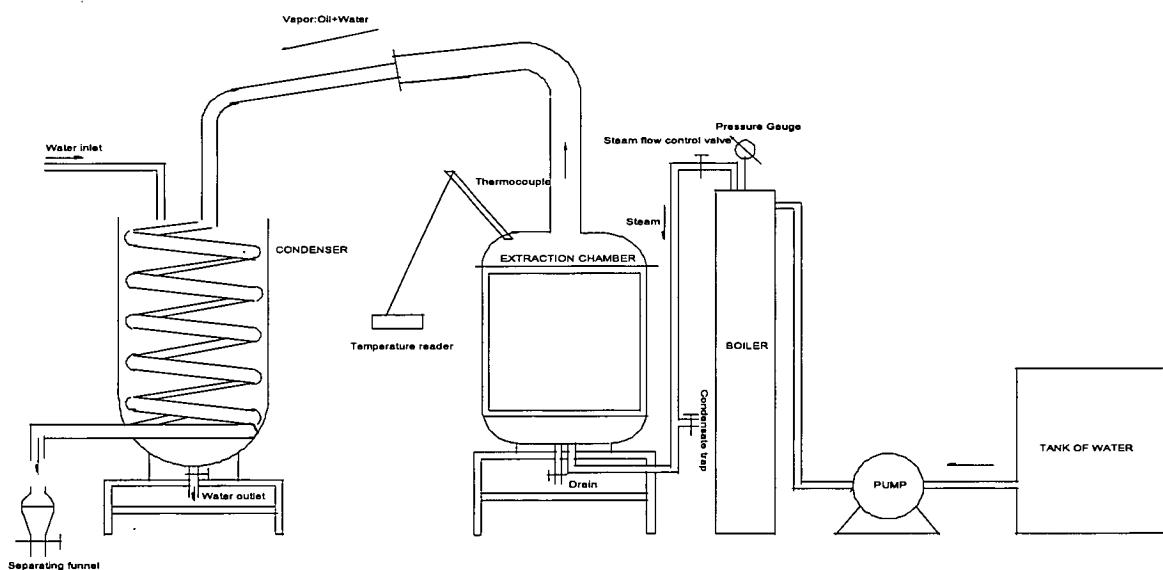


Figure 4.2: Schematic representation of the steam extraction equipment

This type of experimental setup is classified as batch steam distillation equipment which essentially consists of a static bed. This batch equipment possesses advantages of the ease of operation, simple construction and relatively low capital construction cost.

The experimental equipment consisted of the following major individual components

- *Tank:* The purpose of this device was to contain the water supply to the boiler.
- *The boiler and boiler pressure relief valve:* The boiler (Prothem) with a 15kW capacity was connected to three phase power. It consisted of a pressurized air tight stainless steel container and could contain up to ten litre of the water. This was used to generate steam for stripping the plant material, which was supplied via 12.5 mm galvanized pipe. The boiler pressure relief valve was a safety device to prevent it from exceeding a pressure of 1000 kPa, the safe limit of the equipment. If the pressure reached 1000 kPa, the relief valve was intended to open and hence prevent any further pressurization of the boiler.
- *Pressure gauge:* The purpose of this device was to measure the pressure inside the boiler which in effect was the pressure of the steam generated in the boiler.
- *Steam flow control valve:* This device was used to control the flow rate of the steam passing from the boiler, allowing pressurizing of the boiler and its contents.
- *Condensate trap:* This was a process control device which trapped and collected any possible condensate from the stripping column which could potentially collect in the steam line and hence block the steam passage from the boiler to the extraction chamber.
- *Extraction chamber:* This served primary as a container for the plant material, and as a vessel in which the steam contacts the plant material and vaporized its Essential Oils.

It consisted of a cylindrical container manufactured in stainless steel with a diameter of 60 cm, 70 cm high, and was equipped with a removable cover which could be clamped

upon the cylindrical section. On the top of the cylindrical section a pipe was attached to lead the vapours to the condenser.

The bottom of the extraction chamber was provided with a drain valve sufficiently wide so that any water condensing within the charge and dripping to the bottom could be drawn off in the course of distillation. (This drain valve also served as an outlet for the wash water, when the still was cleaned). At the top of the extraction chamber, we installed a thermocouple to measure the temperature at different depths.

- *Condenser:* A coil flow condenser was used to convert all of the steam and the accompanying oil vapours from the top of the extraction chamber into liquid. Water was fed to the overhead reservoir and this permitted the water to trickle over the entire length of the condenser tubes. It will be noted that the condenser tubes were all sloped downward slightly, to insure proper drainage of the condenser oil and steam. The cooling medium used in this device was cooling water drawn from a running tap.
- *Pump* (EBARA) Type:CDxHM/A90/10

A centrifugal pump was used to pump water from the tank to the boiler.

The specifications of the pump are as follows: 0.75kW 20 $\mu$ F Hmax =32m Hmin=19.5m RPM=2840 Phase=1 Kwabs =1.2 Q=20-110l/min Hz=50 P55.

#### **4.3.2 Hazop analyses of the experimental equipment and procedure**

A high level concentration of pure essential oils has known to be extremely toxic, and hence appropriate safety precautions were taken before and during the experiment. The experimental work was performed in a well aerated environment. Safety goggles, laboratory coat, closed

shoes, face gas mask and rubber hand gloves, were worn when carrying out experimental work to avoid contact and exposure to the oils produced.

As the equipment produced and was operated with live steam, proper attention was given to any steam that might have come off from the apparatus. Steam leakages from the apparatus were prevented by use of vacuum seal greases on all connecting joints, ensuring that the fittings were both steam and air tight. Precautionary measures were also taken when opening the boiler at the end of each run to avoid burns that might have arisen from contact with the steam and hot water that remained inside the boiler. It was therefore considered advisable to ensure that the boiler had cooled down before it was touched and opened. Initially, at the start of a batch, the material was placed into the extraction chamber. The boiler was then filled with approximately 5 to 10 litres of tap water, which amounted to about three quarters of its total maximum capacity. It was considered important to note the time at which the temperature reached the desired level to start the extraction process as indicated by the steam boiler pressure gauge. This was to avoid the possibility of vaporization of the essential oils in plant material due to the heating effect prior to commencing of the steam extraction procedure.

A coil flow condenser system was set-up at the exit of the extraction chamber to condense the steam and oil extracts as they left extraction chamber prior to being collected at the exit of the condenser system. When the boiler pressure reached desired operating conditions, extraction was commenced in the chamber by half opening the steam flow valve which released the steam to flow into the steam line and eventually to the extraction chamber. The half valve opening position was found to be the best operating position for this process in test runs. The exit line of the condenser was observed for the first condensate drop into the flask held in position, at which point a stop watch was started. The time the first drop is collected was taken to be the initial extraction time of the oil extracted from the plant material.

#### **4.3.2.1 Boiler**

The boiler was designed to reach a maximum safe operating gauge pressure of 1000 kPa. The boiler had to be handled with great care to avoid burns as it was quite hot.

In addition, it was not recommended that the boiler be operated at a pressure greater than the maximum safe operating gauge pressure as the maximum equipment mechanical safety pressure was unknown. When the level control in the boiler is activated and water is added to the boiler, the temperature drops and steam flow is temporarily reduced. The fresh water could only be added until the top level setting was reached.

#### **4.3.2.2 Steam extraction process procedure**

Steam was generated using a 15 kW boiler, capable of producing 15 kg steam per hour, connected to three phase power. Galvanized tubing (12.5 mm), fitted with an outlet and steam flow control valve, was used to transfer steam to the base of a stainless steel extraction chamber. The extraction chamber, a large pot with a stainless steel jacket, sealed with a rubber O-ring and clips, was 60 cm in diameter and 70 cm high.

The sample was placed in a mesh basket, which was positioned, in a slightly elevated position, in the extraction chamber. After passing through the plant material, steam was released through the top of the jacket into a condenser coil, constructed from 32 mm stainless steel tubing. The extraction chamber was connected to the cooling tower via a Quick coupling. The condenser coil was immersed in a stainless steel drum, which was cooled by passing running water, with a hose, through the drum. An outlet, at the top of the drum, allowed the hottest water to run out to be

replaced by cooler water. A second outlet, at the base of the cooling drum, was used to drain water from the drum, when necessary.

Steam was used as the stripping medium for the extraction of eucalyptus essential oil from the plant material using the experimental equipment show in Figure 4.2.

Each of the samples prepared as mentioned in section 4.2 of this chapter were processed individually as described below in three separate batch samples.

Firstly, the extraction chamber was checked to make sure that no other substance was present which could contaminate the oil extracted.

Secondly, during the processing of each batch, the material was placed into the extraction chamber (see Figure 4.2).

Thirdly, the extraction chamber was closed tightly to prevent any leakage.

Steam distillation commenced 24 h after harvest; by which time the sample of plant material was placed in the weighed sample holder (extraction chamber). The distillate was collected using 250 ml separating funnels at a water flow of approximately 100ml/min. For the first sample, the working pressure was 100kPa and after 6 minutes we collected the first drop of mixture (water+oil). The batch was discontinued once the flow of steam from the boiler ceased. It is important to note that when the pressure reached the respective desired operating condition, stripping was commenced by opening the steam flow valve which released the steam to flow into the steam line and eventually to the extraction chamber. The exit line of the condenser was observed for the first condensate drop in the collector, at which point a stop watch was started. The time the first drop was collected was taken to be the initial extraction time of the oils from the plant material.

Ten weighed empty decanters labelled 1 to 10 were set in position to collect the condensate at 10 minutes intervals once the first condensate drop was observed. The thermocouple reading in extraction chamber was recorded at the different time intervals while the experimental work

progressed. At the end of each experiment extraction run, all the decanters were capped and set to stand on decanter rack ready for their contents to be analysed both qualitatively and quantitatively as will be discussed in the following section.

The system was switched off and the contents (leached plant material) of the extraction chamber were emptied. The equipment had to be cleaned each time before the next run, by running steam through it to ensure that all the extracts which could have condensed on the equipment were removed to avoid cross contamination with the extracts of the runs that followed.

Oil samples extracted were stored in a dark glass bottles with suitable caps in cool place away from light.

#### 4.4 Condensate isolation and characterization of the oils extracts

##### 4.4.1 Isolation of the oil extracts from the steam condensation

Essential oil extracts and water condensate are known to have different densities and also form an immiscible two liquid phase mixture at low room temperature conditions. The separation of the essential oils from the condensate hence utilizes this density and immiscibility advantage for the two to be isolated from each other. This phenomenon is illustrated in Figure 4.3 in which the oil extract is shown to float on the water layer due to being less dense than water.

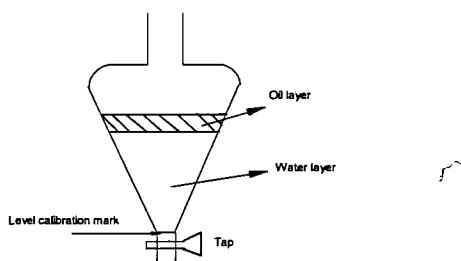


Figure 4.3: Schematic representation of the separating funnel

As shown in Figure 4.3 a calibration mark acted as the reference point during the emptying and determination of the mass of the decanter contents.

In separating the water from the oil, the water layer was carefully run out from the bottom of the decanter by opening the tap until its meniscus was just at the calibration mark. The contents that remained inside the decanter were the oil layer and the water between the tap bridge and the bottom of the calibration mark.

The sample was placed in the apparatus and deionised water was added. A solvent mixture (50ml) of n-pentane and diethyl ether (Column distilled, 1:1, v/v, Merck, Darmstadt, Germany) was used as an extractant. The extract thus obtained was dried over anhydrous sodium sulphate and weighed on the electronic analytical balance. The yield of extraction is defined as the ratio of the mass of essential oil collected after evaporation of the solvents in a rotavapor and the mass of leaves dry matter. A drying oven at 70°C was used to evaporate solvent and determine the dry matter. The sample of oil was transferred into a dark sample bottle and kept a temperature of 4°C for further analysis.

#### **4.4.2 Quantitative characterization of the Eucalyptus Essential Oils extracts**

The quantity of the Eucalyptus Essential Oil extracts was determined by use of an XP-300 top loading electronic analytic balance, which determined their mass within accuracy of 0.001g.

The weighing procedure was a simple technique whereby the wet mass of the empty decanter was first determined by filling it with oil saturated water and emptying it until the level calibration mark. This technique was repeated five times to determine the reproducibility and level of accuracy and hence to have a measure of the human error associated with the decanting technique. The average mass of each set of result was taken as the wet mass of each empty

decanter, while the associated error deviation could be determined. After the wet mass of the decanter had been determined, the combined mass of the oil and water collected during each run in the decanter was determined using the electronic analytic balance. Finally the water was decanted off to enable the total mass of the essential oil extracts to be determined.

#### 4.4.3 Qualitative characterization of the Eucalyptus Essential Oil extracts

The composition of the Eucalyptus Essential Oils extracts was analysed using a CP-3800 Varian Gas Chromatograph (GC) unit. Gas Chromatography involves a sample being injected and vaporised onto the head of a chromatograph column. The sample is then transported through the column by the flow of an inlet carrier phase, whereas the column itself contains a stationary liquid phase which is absorbed onto the surface of an inert solid. Figure 4.4 show a schematic diagram of a gas chromatograph.

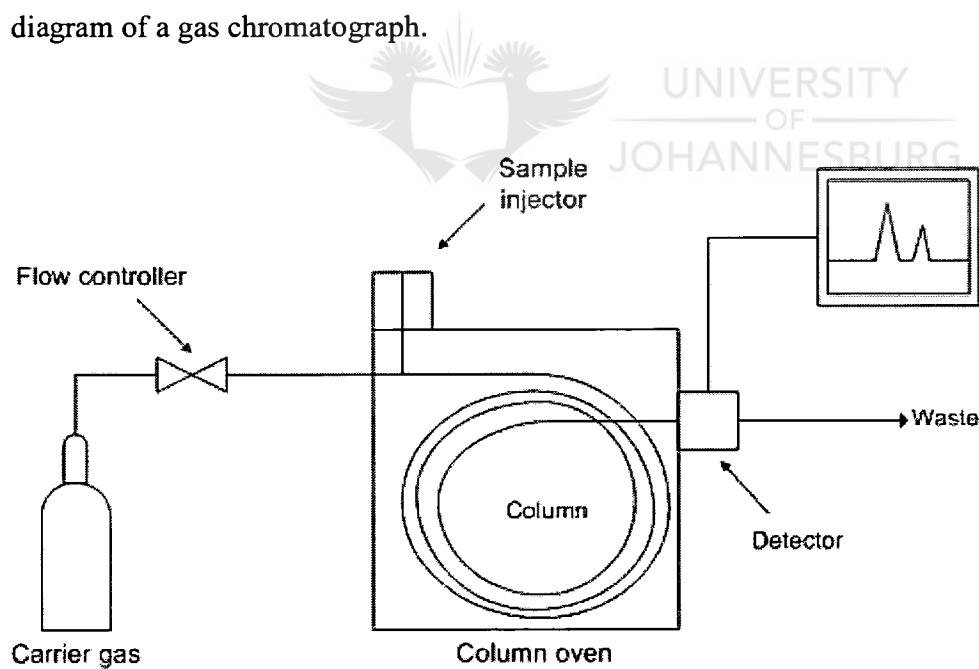


Figure 4.4: Schematic diagram of gas chromatograph [Kasumba, 2003]

A Gas Chromatograph uses a flow through narrow tube (column), through which different chemical constituents of a sample pass in a gas stream (carrier gas) at different rates depending

on their various chemical and physical properties and their interaction with a specific column filling (stationary phase).

The Gas Chromatograph was filled with a fused silica capillary column, split-less injector, a vapour sampling loop and a Flame Ionization detector (FDI). Chemically inert Helium gas was used as both a reference and carrier gas during sample analysis.

The Gas Chromatograph was set to the determined optimum operating condition tabulated in Table 4.1.

**Table 4.1: GC set optimum operating conditions**

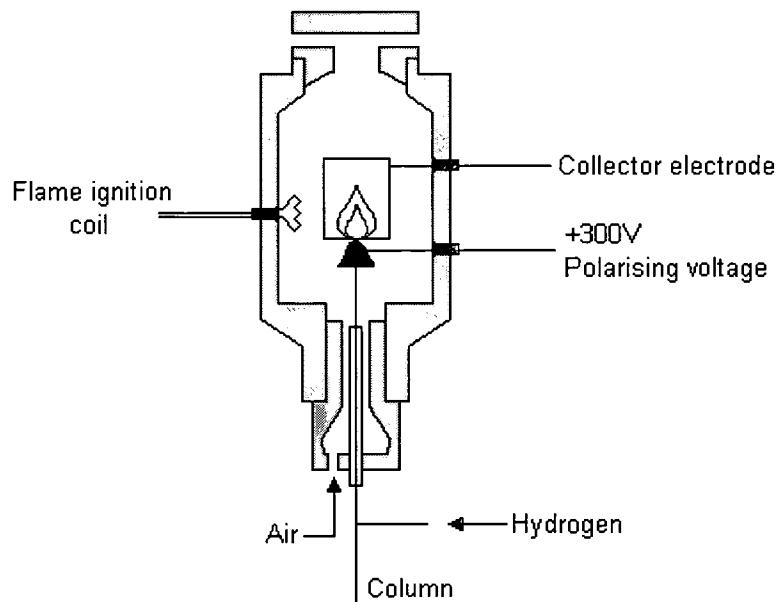
Variable	Value
Injector	Split/ split less used with 1:30 split ratio
Carrier gas	Helium (Afrox, Instrument grade: 99.999%) Flow rate: 4.5ml/min
Detector gases	Compressed air (dry): 300 ml/min Hydrogen (Afrox, Instrument grade: 99.999%): 30 ml/min
Make-up gas	Nitrogen (From liquid nitrogen evaporator; 99.99%) Flow rate: 26ml/min
Analytical column	Phenomenex ZB-5 (cross-linked 5% diphenyl 95% dimethyl siloane)
Column specification	30m; 0.25mm id; 0.25µm Stationary phase layer

Injector temperature	280°C
Over temperature	Initial: 50°C (10 min) Ramp rate: 2.5°C/min Final temperature: 220°C (10 min)
Detector temperature	300°C

A sample of 2.0 $\mu$ l of each of the oil extract samples was collected at different pressures and 100  $\mu$ l of Hexane (vaporised solvent) was prepared as dilute solution due to the sensitivity of the detection methods.

The dissolved sample (1.0 $\mu$ l), dissolved was drawn and injected through the rubber septum at the head of the column for each individual run. A glass barrel syringe was used where the stainless steel needle forms the sample reservoir. The sample was pulled into the needle and expelled by a stainless steel plunger wire protruding from the end of the needle whilst in the empty position. Both the needle and barrel are accurately constructed and since almost all the sample is expelled from the syringe there is a minimum dead volume and sample retention, thus avoiding as far as possible any cross-contamination of samples.

## The Flame Ionisation Detector



**Figure 4.5: The Flame Ionisation Detector [Still, 1978]**

The syringe was fully loaded, carefully checked to make sure that no air bubbles were present, and adjusted to the required injection volume of  $1\mu\text{l}$  for GC analysis. The needle was introduced into the injection port through a self-sealing elastic septum, the sample rapidly expelled and the syringe quickly withdrawn to avoid vaporization of additional sample which might have been retained in the needle. The syringe was regularly flushed out with the samples to be used to avoid cross-contamination of the samples.

The micro syringe needle was placed into a hot injector part of the Gas Chromatograph, where the sample was injected.

## The split / splitless injector

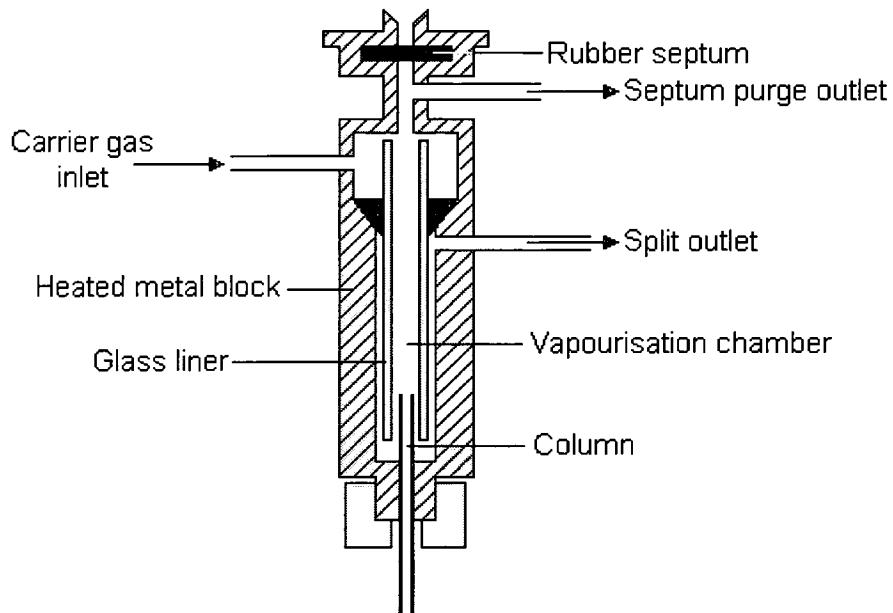


Figure 4.6: The split/ split less injector [Still, 1978]

When the liquid oil extract sample was injected into the column, it was vaporised and mixed with the carrier gas thereby forming a mixture of carrier gas and vaporised solvent. The carrier gas flowed through the injector and pushed the gaseous components of the sample into the Gas Chromatograph column. It was within the column that separation of the components took place. The molecules were partitioned between the carrier gas and the stationary phase within the GC column. As the carrier gas (Helium) swept the entire sample (split less mode) of the sample through the column, this motion was inhibited by the adsorption of the sample onto the column walls. The rate at which a given molecule progressed along the column depended on the strength of adsorption, which in turn depended on the type of molecule and on the stationary phase materials (the function of the stationary phase in the column was to separate different components, causing each one to exit the column at a different times). Since each type of molecule had a different rate of progression, the various components of the mixture were

separated as they progressed along the column and reached the end of the column at the different times (retention times). After components of the mixture had moved through the Gas Chromatograph column, they reached a detector. Components of the mixture would have reached the detector at varying times due to differences in the partitioning between mobile and stationary phases. The Flame Ionized Detector (FID) thereby produced a voltage signal which was captured using the FID chemstation software. The FID chemstation software analysed the output peak areas and converted them into the corresponding compositions of the different components in the sample mixture. The FID basically compared the thermal conductivities of the two gases flowing into it which was the reference gas (pure carrier gas) and the column effluent (pure carrier gas and sample component mixture).

The detector sent a signal to the chart recorder which resulted in a peak on the chart paper. The area under each GC peak is hence proportional to the amount of a specific component in a mixture generating the signal.

A detector was used to monitor the outlet stream from the column; thus, the time at which each component reached the outlet and the amount of that component could be determined. Substances were identified electronically (qualitatively) by the order in which they emerge from the column and by the retention time of the sample in the column.

Separation of the components of the mixture occurred in the column compounds were differentially retained in the stationary phase to reach the detector at different times to produce a set of peaks along the time line. The detector response was sent to a computer system where the progress of the sample was monitored on the computer in graphical form that displayed detector responses as a function of run time.

From typical Gas Chromatograph peaks, it is impossible to tell which peak represents which component, thus it is necessary to determine the retention times of the component in order to relate them to their corresponding areas. The retention time is the time taken from when the

sample is injected into the column to when it reaches the detector for the component peak to be detected and produced. Each of the samples collected were injected separately to identify the different quantities of the components contained in them. Nearly 220 constituents were identified eight different major peaks were detected overall in all the injected samples and the corresponding areas recorded accordingly. The areas of the minor peaks were ignored and hence the results were corrected and normalised to that of the eight major peaks in order to determine the corresponding fractions in each injected sample. The area percentages recorded represent the compositions of the components in each corresponding extracted sample. Literature [Hammersmith, 1995] reports that Pinene, Globulol, Limonene, pinocarvone, Myrcene, Camphene, Citronellal and Cineol seem to be the major components of Eucalyptus Essential Oil, as the finger prints of these specific Eucalyptus Essential Oils were unknown for the CP-3800 Varian Gas Chromatograph used, the main peaks were numbered and no attempt was made to identify the actual components to match what is reported in the literature [Hammersmith, 1995]. Typical chromatograms of the Eucalyptus leaves extracts are shown in Appendix C.

#### 4.5 Conclusion

The experimental apparatus that was used to study the effect of various operating conditions on the yield and quality of essential oil extracted by steam distillation has been presented together with the experimental method that was followed. An electronic top loading analytical balance and Gas Chromatograph were used to obtain and analyse both the experimental results for the quantity and quality of the oil extracts respectively. The experimental raw data obtained from these experiments for both the quality and yield of essential oil was determined and are summarised and shown in Appendix A.

## **Chapter 5**

### **Results and Discussions**

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#### **5.1 Introduction**

The effects of operating parameters such as pressure and temperature on the quantity and quality of essential oils extracted by means of steam extraction were studied using Eucalyptus leaves as the plant material. The aspects of the results studies here include both quantitative and qualitative analyses obtained by use of an electronic analytical balance and Gas Chromatography respectively as has been discussed in Chapter 4.

To thoroughly investigate the effect of operating parameters on the yield and quality of oil extracts, the focus area in the analysis of the results was directed towards the following listed points:

- Quantity of the oil extracted (yield)
- Quality of the oil extracted (oil extract composition)
- Mathematical modelling of the extraction process

The quantitative, qualitative and mathematical modelling results are reported in tabulated format in Appendices A to C.

## 5.2 The quantitative analyses of the oil extracts

The literature indicates that the total yield depends on extraction pressure and temperature. Designed experiments were carried out to map quantitative effects of these parameters. The yields of the extraction is calculated from the relation between the essential oil mass obtained and the raw material mass used in the extraction ( $Y = \text{extract/raw material, \%}$ ). The yield curve is constructed from oil mass extracted in relation to the amount sample used in the extraction. In this work we used 20kg of leaves.

The pressure in the boiler for consecutive tests was set at 80kPa (minimum setting), 100kPa and 200kPa. This resulted in extraction chamber temperatures of 90°C, 97°C and 99°C respectively. Higher pressures would have resulted in higher temperatures that would destroy components in the essential oil. The yield (quantities) of the oil extracts from every 20 kg of Eucalyptus leaves subjected to the operating parameters were obtained and are shown in Figure 5.2.

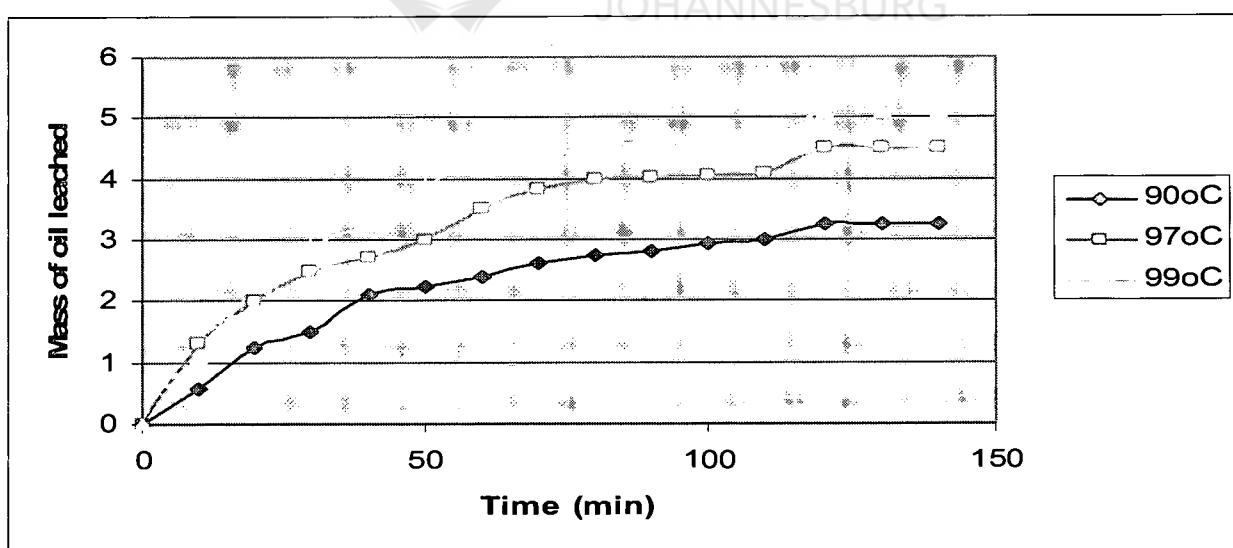
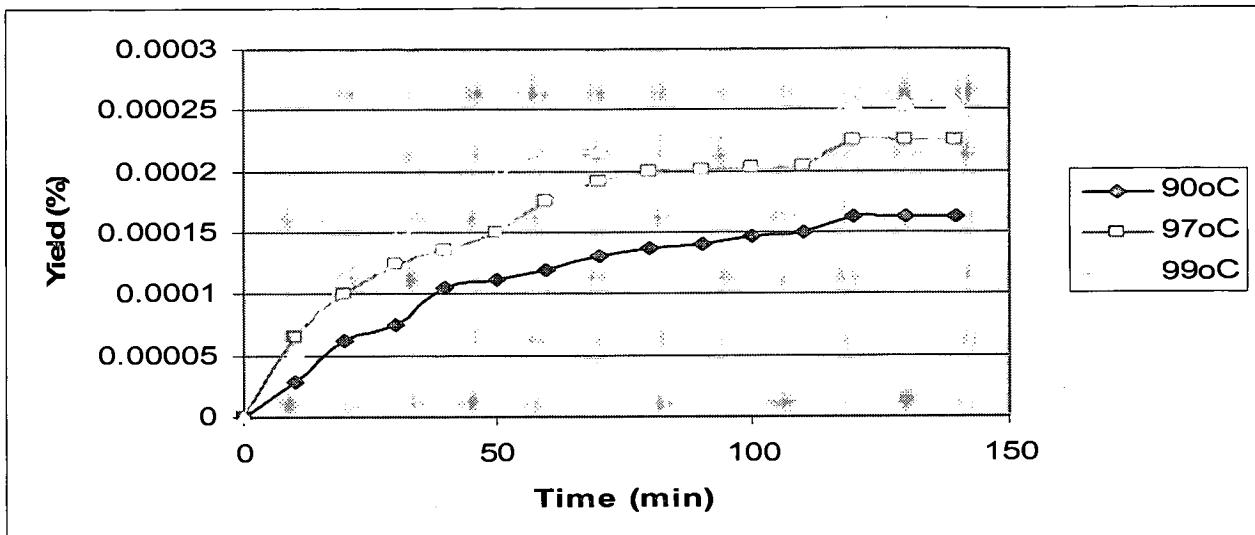


Figure 5.1: Cumulative mass of oil extracted



**Figure 5.2 Yield curve of the Eucalyptus essential oil**

Figure 5.1 above shows the results for the cumulative mass of oil extracts plotted against the extraction time. From this graph it can be observed that the oil extracted at 99°C provide the highest overall cumulative mass after 120 minutes extraction period followed by the oil extracted at 97°C. It is also evident that there is no more oil coming off at all temperatures after 120 minutes mark as all cumulative curves appear to flatten off beyond this point. From this result it can be noted that the optimal extraction time for all temperatures is about 120 minutes which ensures the recovery of all the extractable oil components. This would mean that extractable oil decreases as the temperature decreases. (See the modelling results- the decreased recovery may on the other hand only be the temperature effect on the diffusion coefficient).

The quantitative study of the oil extracts can be used to give an insight on the effect of the temperature on oil extraction during the process. The quantitative results are bound to be helpful in studying the effects on mass of oil extracted.

### **5.3 The qualitative analyses of the oil extracts**

This section is intended to study and analyse the effect on the quality of the oil extracts as a result of the operating parameters employed on the plant material (leaves). The use of the gas chromatograph makes possible to separate the volatile components of Essential Oils and to determine the amount of each component present. The components in the sample get separated in the column because they take different amounts of time to travel through the column depending on how strongly they interact with the stationary phase. As the components move into the column from the injection port they dissolve in the stationary phase and are retained. The rate at which compounds move through the column depends on the nature of the interaction between the compound and the stationary phase. The GC was equipped with flame ionization detectors (FIDs). FIDs use an air/hydrogen flame to pyrolyze the effluent sample. The pyrolysis of the compounds in the flame creates ions. A voltage is applied across the flame and the resulting flow of ions is detected as a current. The number of ions produced, and therefore the resulting current, depends on the flame conditions and the identity of the molecule in question. In other words, the detector (converted from current to voltage) is sent to an integrating recorder that plots, stores, and analyzes the data (see Appendix C). The detector voltage (y-axis) is plotted as a function of time (x-axis). Each peak corresponds to a separate component. The time it takes for a given peak to appear after injection is called the retention time. Eight different major peaks were detected overall in all the injected samples and the corresponding areas recorded accordingly. The areas of the minor peaks were ignored. The area percentage of each of the eight detected components of Eucalyptus oil extracts was obtained and recorded in Appendix A. It was assumed that the area percentage approximated the mass composition of the components in the oil extracted sample. The different peaks represent the different components that are found in this species of

Eucalyptus oils and are shown with their corresponding composition in Table A.2. The sole purpose of gas chromatographic investigation was to indicate the difference in the profile of volatile compounds; there was no prior intention to identify them.

**Table 5.1: A table summarizing the 8 components**

Retention time	Area	Area (100%)	Height
2.776	27420.2071	18.33	3240.263
2.968	26183.5096	17.50	2818.963
3.238	47050.8092	31.45	4735.439
3.623	21451.2868	14.34	3128.712
4.085	5189.7176	3.47	1047.098
4.205	2563.3858	1.71	630.392
5.558	2011.3796	1.34	541.894
9.688	8452.5883	5.65	1482.158

#### **5.4 The mathematical modelling of the oil extraction process**

The Fick's law of diffusion was used because it is the more fundamental, straight forward way to model diffusion processes and can be used to solve for the diffusion coefficient D. Fick's first law was used assuming quasi- steady-state diffusion i.e., when the concentration within the diffusion volume does not change significantly with respect to time. ( $J_{in} = J_{out}$ ). The first law deals with the passage of a gas through a membrane.

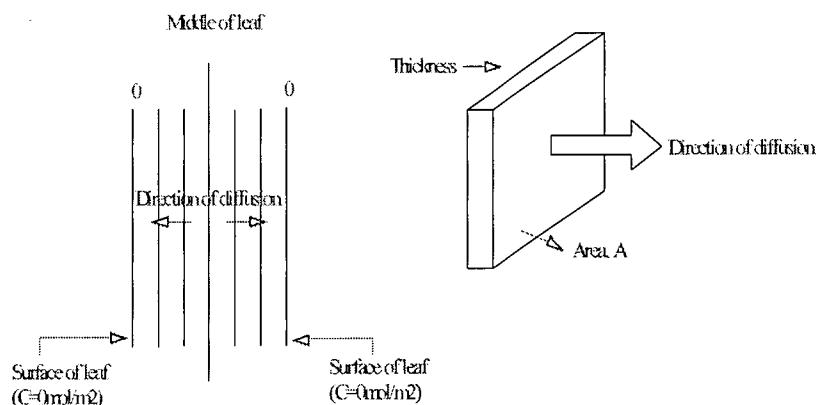
$$\text{In one (special) dimension, this is: } J = -D \frac{\partial C}{\partial x}$$

5.1

D is the diffusion coefficient of the average oil molecule (see results). It is assumed that the oil has mostly the same composition so that a single diffusion coefficient will suffice in the modelling.

#### 5.4.1 Numerical method and problem description

A mathematical model defined as the numerical method needed to be developed to describe the oil extraction process and mechanism during the steam extraction of Eucalyptus leaves. This model was used to fit the experimental data for the variation of oil leached with time and hence compare the diffusion coefficients of the oil extraction for different temperatures. The flux of diffusing atoms,  $J$ , is usually used to quantify how fast diffusion occurs. The flux is defined as the mol flux of atoms diffusing through unit area per unit time, (e.g., mol/m<sup>2</sup>-second).



**Figure 5.3 Schematic representation of diffusion in the leaf**

The minus sign in the Equation 5.1 means that diffusion is down the concentration gradient. The leaf was divided into sections of thickness  $\Delta x$ . The concentration of oil at the surface was assumed to be  $0\text{mol/m}^3$ . If  $\Delta C$  is the concentration difference between two sections, Fick's first law (the diffusion flux along direction  $x$  is proportional to the concentration gradient) may be written as:

$$J = \frac{\Delta n}{A\Delta t} \quad 5.2$$

$$\frac{\Delta n}{A\Delta t} = -D \frac{\Delta C}{\Delta x} \quad 5.3$$

After a short time  $\Delta t$ , the difference in the number of moles diffusing out and in a given section respectively are given by

$$\Delta n_{out} = -A\Delta t D \frac{\Delta C_1}{\Delta x} \quad 5.4$$

$$\Delta n_{in} = -A\Delta t D \frac{\Delta C_2}{\Delta x} \quad 5.5$$

Where:

$\Delta C_1$  is concentration difference in the section of the leaf and the next section in the direction of the edge of the leaf.

$\Delta C_2$  is concentration difference in the previous section of the leaf and the current section.

$\Delta n_{out}$  is the number of atoms diffusing out of the leaf

$\Delta n_{in}$  is the number of atoms diffusing in the leaf during time interval  $\Delta t$ .

Net loss of  $n$  (the number of atoms diffusing through the area of leaf)

$$\Delta n_{out} - \Delta n_{in} = -A \frac{\Delta t}{\Delta x} D (\Delta C_1 - \Delta C_2) \quad 5.6$$

Drop in concentration

$$\frac{\Delta n_{out}}{\Delta x} - \frac{\Delta n_{in}}{\Delta x} = -A \frac{\Delta t}{\Delta x^2} D(\Delta C_1 - \Delta C_2) \quad 5.7$$

$$\frac{\Delta n_{out}}{A \Delta x} - \frac{\Delta n_{in}}{A \Delta x} = -D \frac{\Delta t}{\Delta x^2} D(\Delta C_1 - \Delta C_2) \quad 5.8$$

$$C_{out} - C_{in} = D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2) \quad 5.9$$

$$C_{out} = C_{in} - D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2) \quad 5.10$$

$$C = C_o - \frac{D \Delta t}{(\Delta x)^2} (\Delta C_1 - \Delta C_2) \quad 5.11$$

From this equation, we can generalise:

$$C_n = C_{n-1} - D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2) \quad 5.12$$

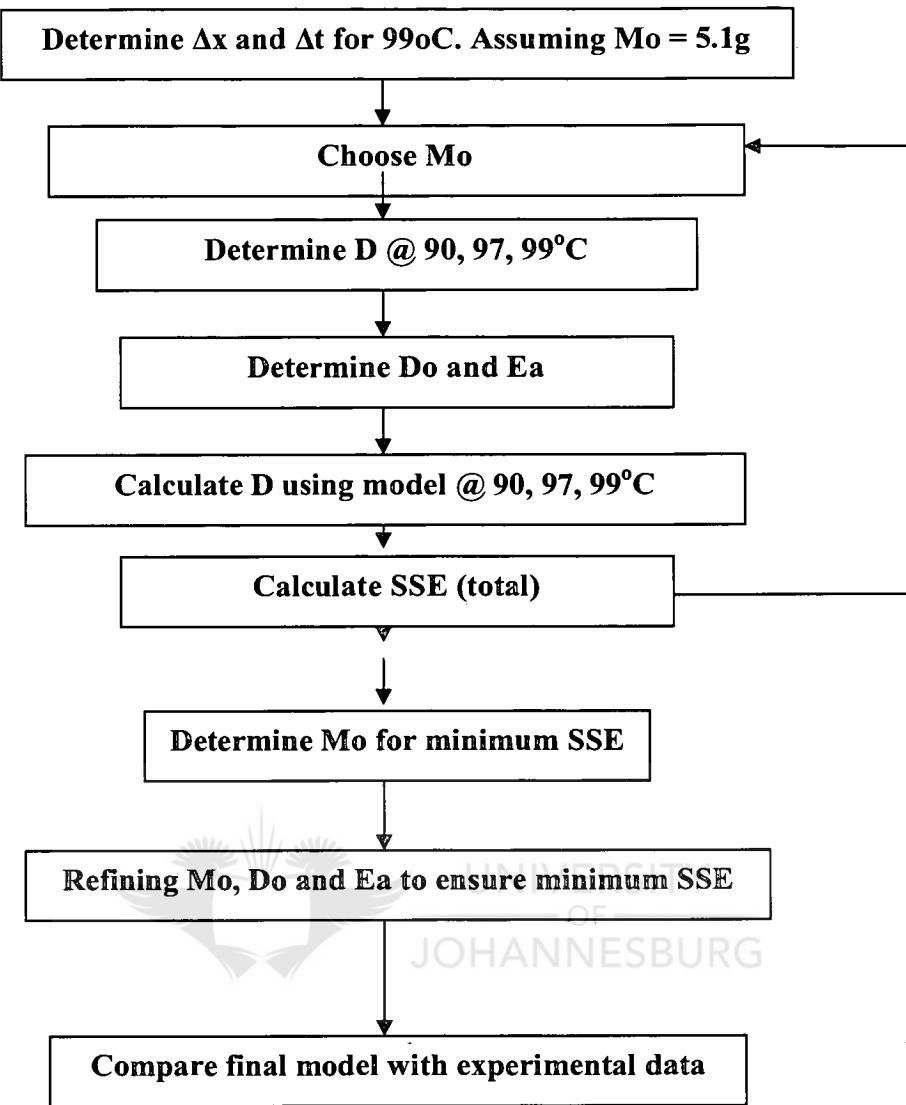
Hence one can calculate the concentration in a given section of leaf ( $C_n$ ) after a time  $\Delta t$  has elapsed using data from the previous time interval.

#### 5.4.2 Model development

The numerical model for the extraction will enable better understanding of the extraction mechanism of essential oils from eucalyptus leaves.

The numerical modelling is also very important in the scale up process from laboratory work to actual industrial process as well as providing an insight into the extraction mechanism. The mass transfer model was used to simulate experimental conditions obtained for steam extraction of Eucalyptus essential oil at 150kPa and 99°C.

The model development is represented by a simple schematic diagram (Figure 5.4) and the steps are listed below from sub-section 5.2.2.1 onwards:

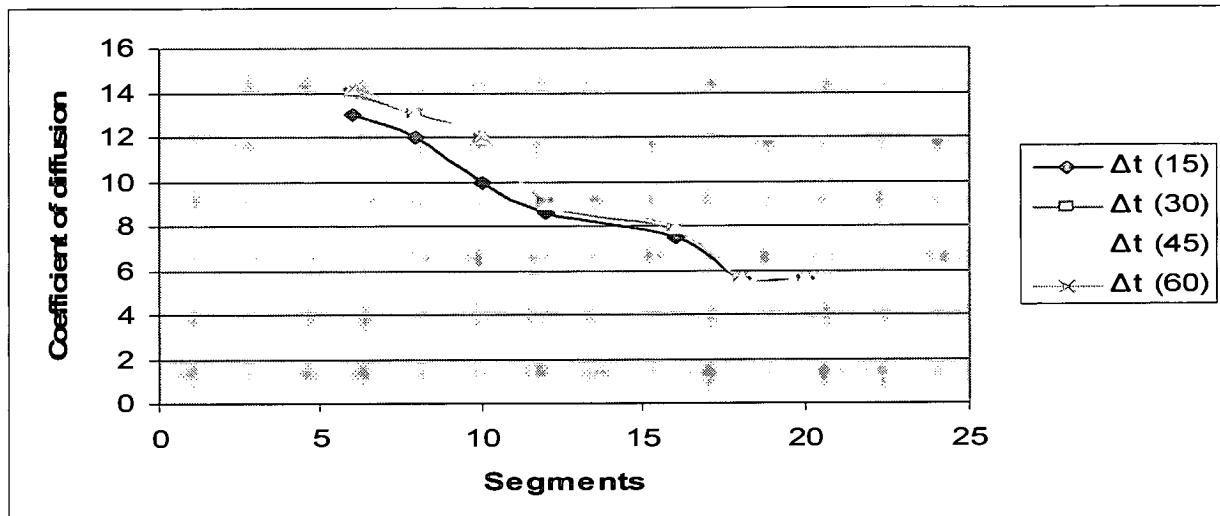


**Figure 5.4:** Schematic diagram of modelling steps

#### 5.4.2.1 Determining of $\Delta x$ and $\Delta t$

Determination of  $\Delta x$  and  $\Delta t$  for 99°C (as maximum temperature) was done by assuming an initial oil mass in the leaves of  $Mo= 5.1$  kg. In order to determine the required number of segments (determined by the value of  $\Delta x$ ), the number of segments in the spreadsheet were increased until the best fit value for D reached a steady value (see Figure 5.5). It was then deduced that

increasing the number of segments above 18 did not make the modelling more accurate. The same principle was followed to determine  $\Delta t$ . Here  $\Delta t$  was decreased until the best fit value for D became constant (see Figure 5.5), which happened when  $\Delta t$  was 30 minutes.



**Figure 5.5: Determination of required segment number and delta t at 99°C for use in the spreadsheet.**

The equation:  $C_n = C_{n-1} - D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2)$  was used in the spreadsheet shown in Appendix B. It was assumed that the percentage of oil condensate concentration at edge of the leaves was

equal to 0 because mol fraction of oil  $= \left( \frac{n_{oil}}{n_{H_{2o}} + n_{oil}} \right)_{vapour} = \left( \frac{n_{oil}}{n_{H_{2o}} + n_{oil}} \right)_{condensate} = 0.000011$  as calculated from the amount of oil and water collected after condensation. This value can be seen to be close to 0. This since mass of oil and water collected are 5.1 g and 60000g respectively

#### 5.4.2.2 First Choice of Initial mass (Mo)

At first, the initial mass of oil was assumed to be the maximum value obtained in the experiments (5.1g at 99°C). Later on, Equation 5.12 was used in the spreadsheet to determine a more accurate value for Mo (initial mass – see Sub-Section 5.4.2.7).

#### 5.4.2.3 Determining of Coefficient Diffusion at 90, 97 and 99°C

The best fit values of D were then determined for each temperature and the SSE was calculated. In the diffusional model, the parameter D was directly adjusted using the Equation 5.12 with the data from the spreadsheet. This parameter was estimated by minimization of the sum of square errors (SSE) between the experimental data obtained and the prediction from the model.

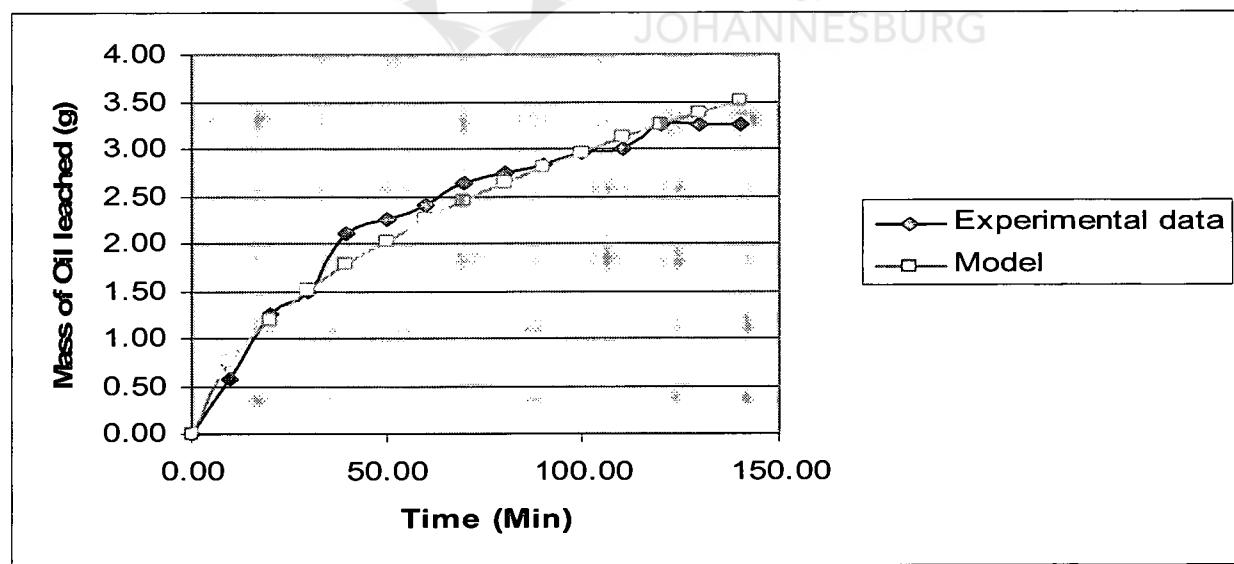


Figure 5.6: Fitting the Theoretical oil extraction model using the best fit D value to 90°C experimental data

Analyzing the Figure 5.6, it was noted that the diffusional model fitted very well the experimental data and the determined value was  $D = 3.00 \times 10^{-12} \text{ m}^2/\text{sec}$  with  $\text{SSE} = 0.360$ .

The same principle was used also in Figure 5.7:  $D = 6.40 \times 10^{-12} \text{ m}^2/\text{sec}$  with  $\text{SSE} = 0.301$  and in Figure 5.8:  $D = 9.30 \times 10^{-12} \text{ m}^2/\text{sec}$  at  $99^\circ\text{C}$  with  $\text{SSE} = 0.404$

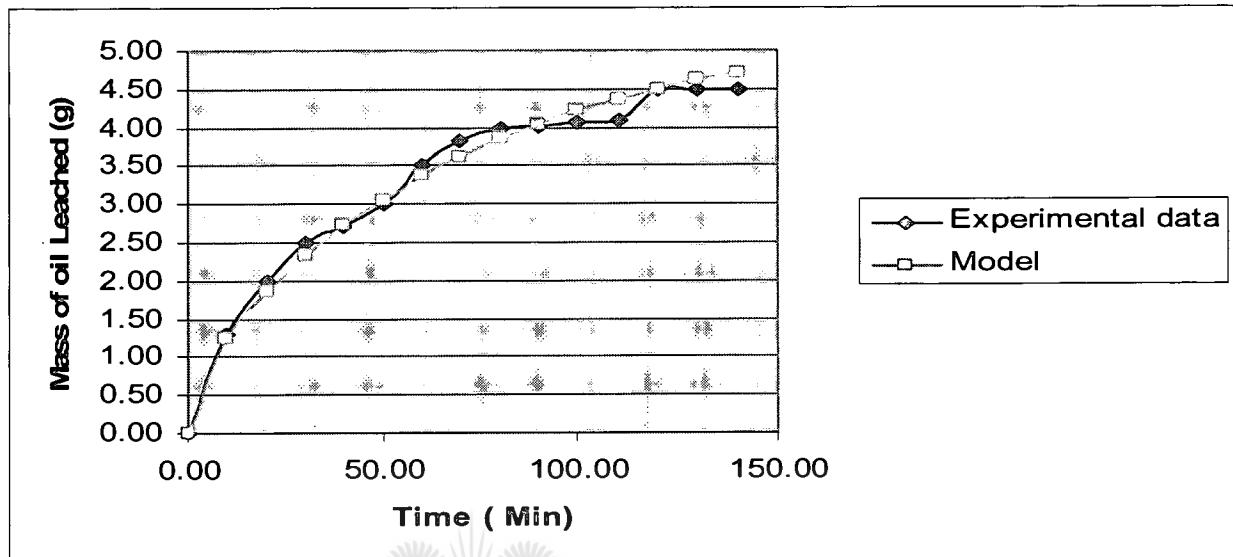


Figure 5.7: Fitting the Theoretical oil extraction model using the best fit D value to  $97^\circ\text{C}$  experimental data

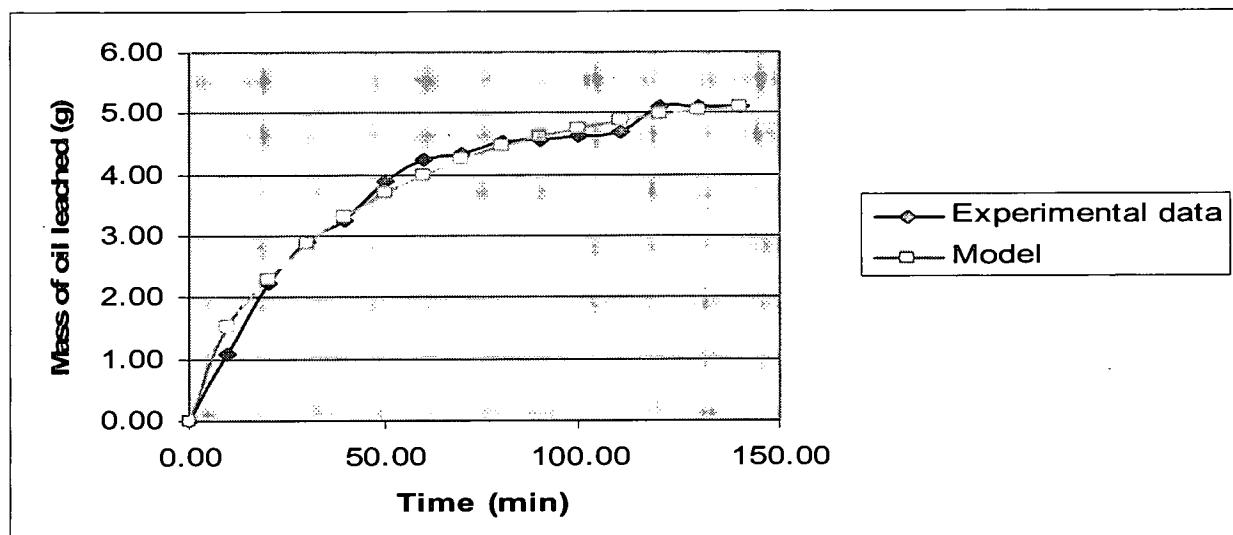


Figure 5.8: Fitting the Theoretical oil extraction model using the best fit D value to  $99^\circ\text{C}$  experimental data

#### 5.4.2.4 Determination of Do and Ea

The diffusion coefficients at 90, 97 and 99°C were employed to determine Do and Ea using the linear regression as illustrated in Figure 5.9. The Activation Energy (Ea) and Maximum Diffusion Coefficient (Do) were determined graphically by using the linear regression from Figure 5.9. Here the Arrhenius Equation:

$$D = D_0 e^{-\frac{E_a}{RT}} \quad 5.13$$

was used, is in its logarithmic form:  $\ln D = \ln D_0 - \left(\frac{E_a}{RT}\right)$ .

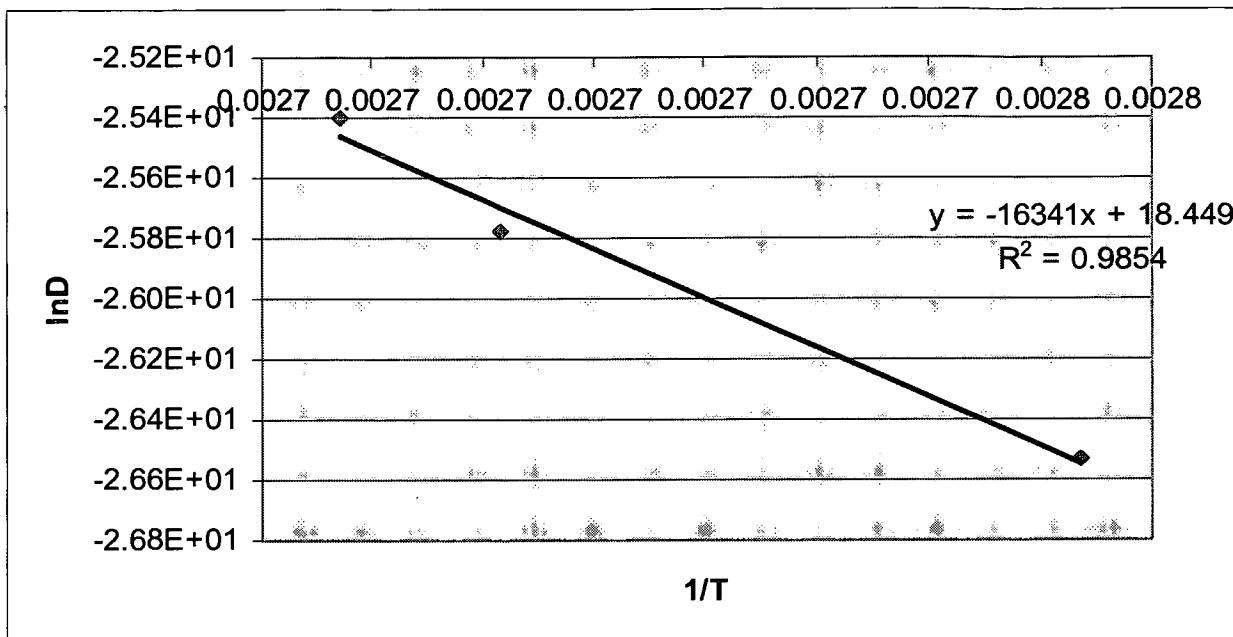
Equation 5.13 shows that the coefficient of diffusion varies with the temperature T, which also means that the quantity  $\ln D$  varies with the quantity  $(1/T)$ . These two quantities  $\ln D$  and  $1/T$  are variables and we had an equation for a straight line.

$$y = b + m x \quad 5.14$$

Thus, to determine the activation energy, we constructed a graph of  $\ln D$  versus  $1/T$ , measured the slope of the line, and then used the relationship. The best fit values (minimizing the SSE) of D from the spreadsheet for the various temperatures were used to calculate the  $\ln D$  values.

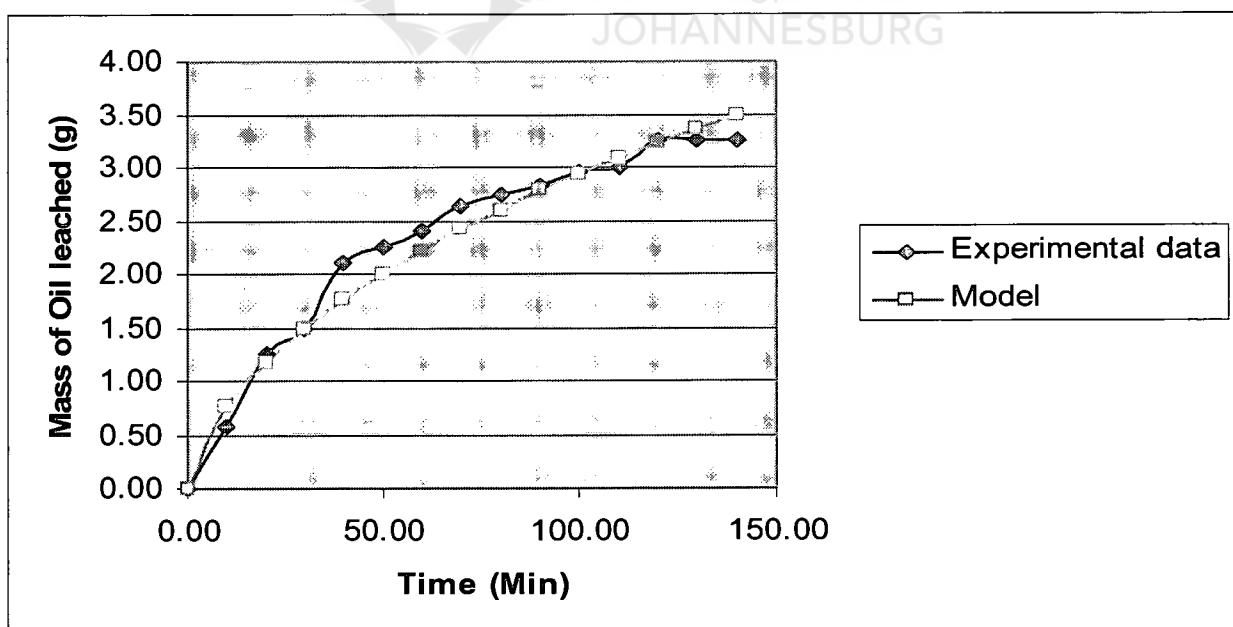
$$\text{Slope} = -\frac{E_A}{R} \quad 5.15$$

Mass (m)	-16340.89513
Activate Energy (kJ/mol)	135.8582021
$\ln D_0$	18.44854
Maximum diffusion coefficient ( $m^2/s$ )	102825214.7



**Figure 5.9: Determination of the activation energy:  $\ln D$  versus  $1/T$**

#### 5.4.2.5 Calculation of Diffusion Coefficient using model at 90, 97 and 99°C



**Figure 5.10: Fitting the Theoretical oil extraction model using calculated  $D$  values using equation 17 to 90°C experimental data**

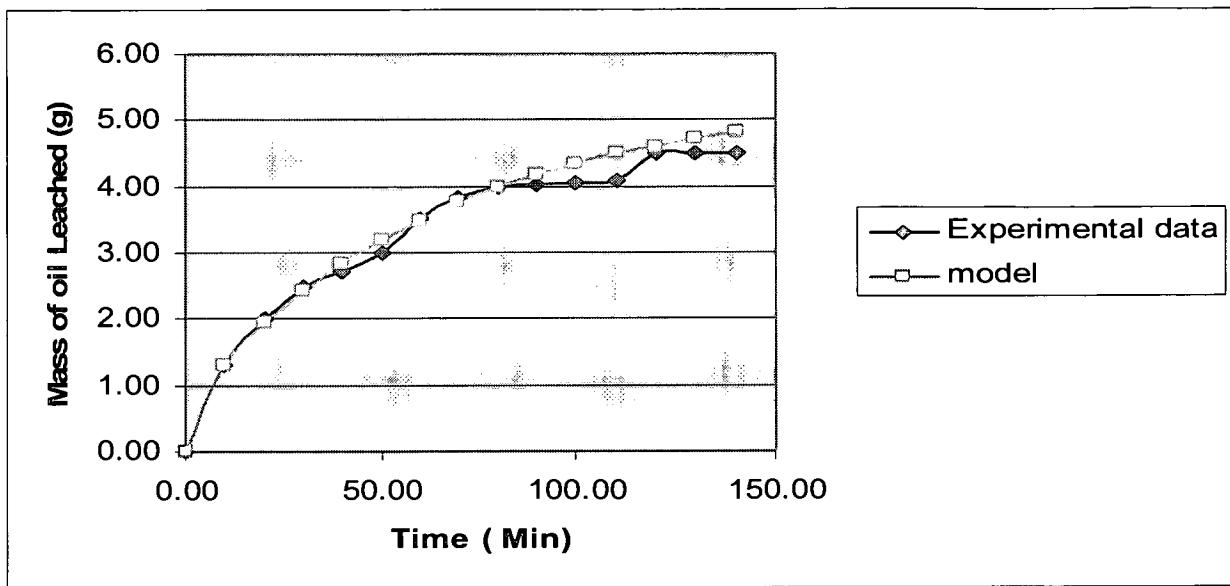


Figure 5.11: Fitting the Theoretical oil extraction model using calculated D values using Equation 5.13 to 97°C experimental data

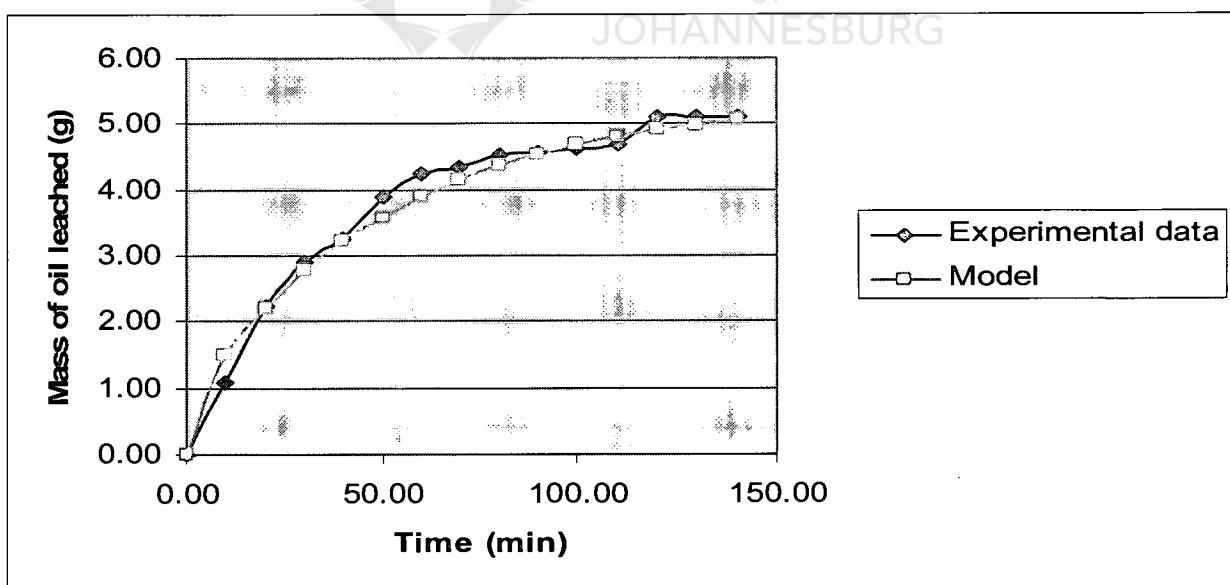


Figure 5.12: Fitting the Theoretical oil extraction model calculated D values using Equation 5.13 to 99°C experimental data

From Figure 5.10 to 5.12, it can be seen that the values for D calculated from the model using Equation 5.13 give a good fit to the experimental data, only slightly less accurate than using the individual best fit values determined for each temperature. The calculation of the coefficients of diffusion using the model at 90, 97 and 99°C was done on the spreadsheet by fitting the experimental data.

The calculated values (using Equation 5.13) for the diffusion coefficient at the various temperatures were:

$$D = 2.93 \times 10^{-12} \text{ m}^2/\text{sec} \text{ at } 90^\circ\text{C}$$

$$D = 6.86089 \times 10^{-12} \text{ m}^2/\text{sec} \text{ at } 97^\circ\text{C}$$

$$D = 8.6986 \times 10^{-12} \text{ m}^2/\text{sec} \text{ at } 99^\circ\text{C}$$

It is largely because T occurs in the exponential function of the Arrhenius Equation that small changes in T can mean large changes in D.



#### 5.4.2.6 Calculation of Sum of Square Errors (Total)

**Table 5.2: Minimum SSE with corresponding D values versus Temperature for Mo=5.4 g**

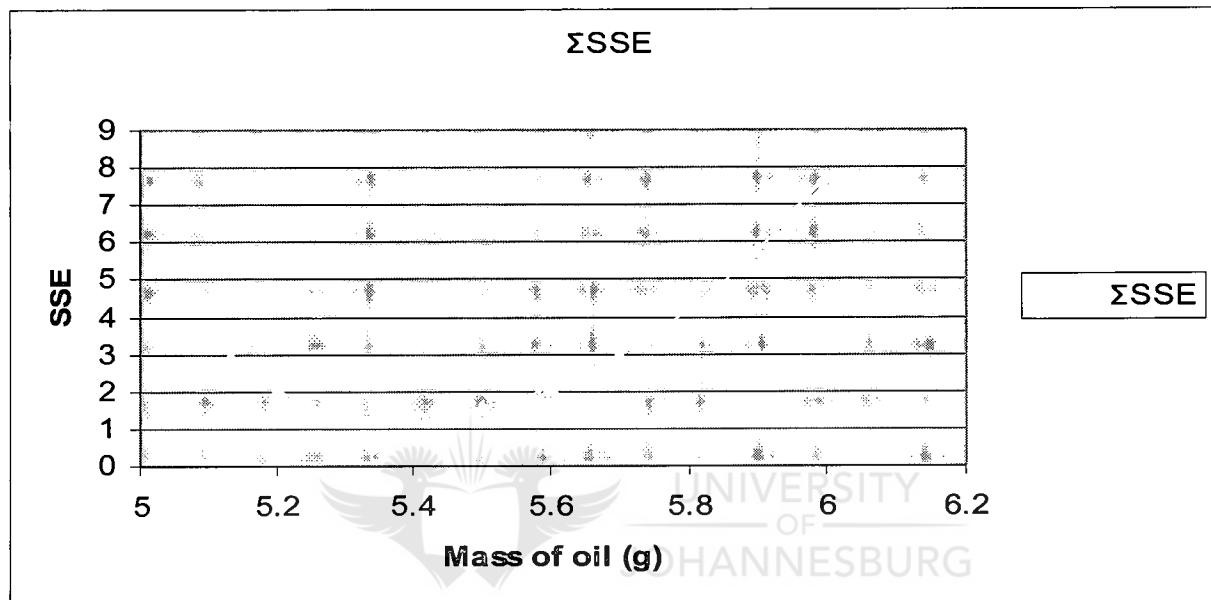
Diffusion Coefficient ( $\text{m}^2/\text{s}$ )	Temperature ( $^\circ\text{C}$ )	SSE
$2.94 \times 10^{-12}$	90	0.382
$6.89 \times 10^{-12}$	97	0.477
$8.74 \times 10^{-12}$	99	0.511
<b>Sum of Square Errors (Total)</b>		<b>1.37</b>

The calculation of SSE (total) was obtained from the spreadsheet by adding the sum of square errors for the different temperatures using the initial mass  $M_0 = 5.4\text{g}$  and the value were calculated in Table 5.2.

#### **5.4.2.7 Determination of Mo for minimum SSE**

Several values of Mo were chosen and the one (5.4 g) that gave the smallest overall SSE was selected (see Figure 5.13).

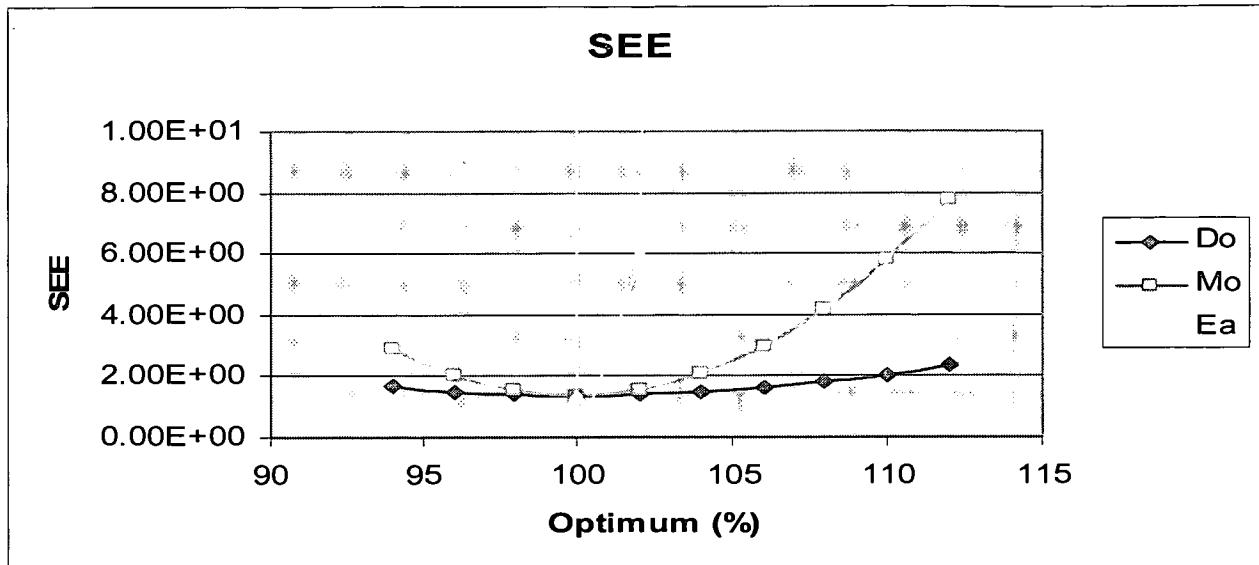
A value of 5.4g for Mo was obtained by minimizing the SSE as shown in Figure 5.13.



**Figure 5.13: Determination of initial mass of oil (Mo)**

#### **5.4.2.8 Refining of Do, Ea and Mo**

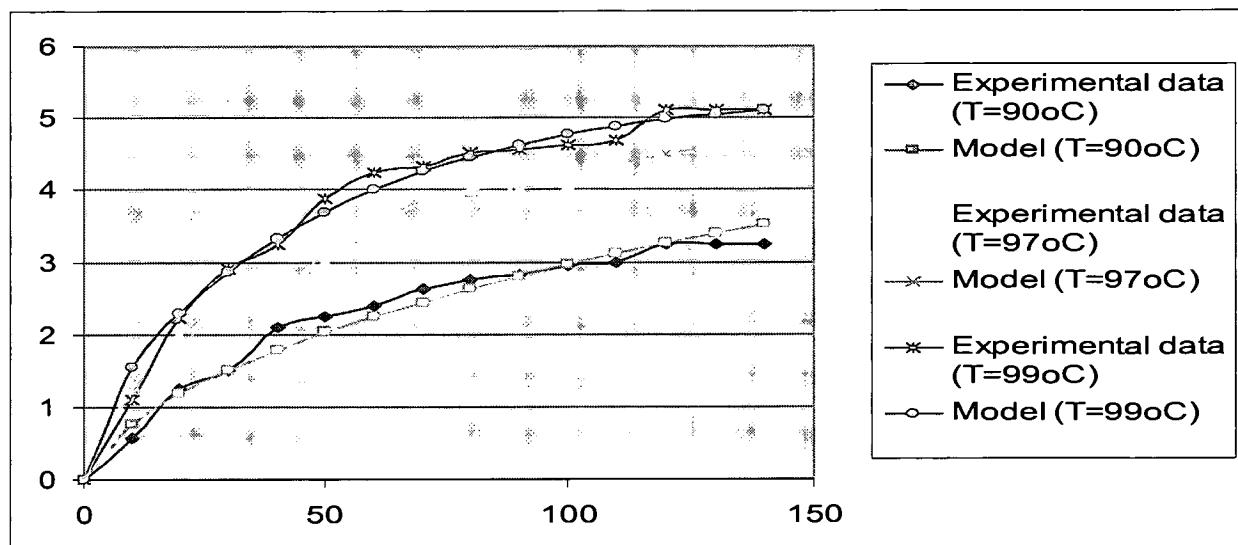
The values of Do, Ea and Mo were changed by small amounts and the effect on SSE was noted. In this way, the absolute minimum value of SSE could be established with the corresponding optimum (refined) values of Do, Ea and Mo. The effect of changing these values one by one by a small amount (94% to 112% of the respective optimum values) is shown in Figure 5.14, where the SSE for all experiments ( $90^{\circ}\text{C} + 97^{\circ}\text{C} + 99^{\circ}\text{C}$ ) is calculated.



**Figure 5.14: Optimization of Do, Mo and Ea versus SSE**

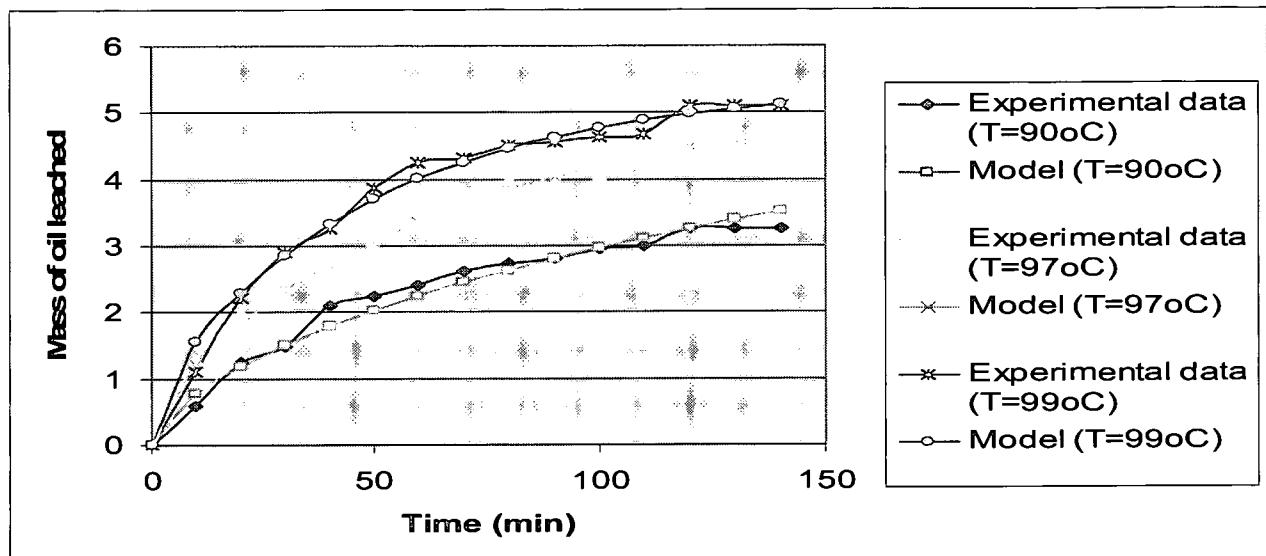
From Figure 5.14 above, the optimum values of Do, Mo and Ea were confirmed by minimizing the SSE. Only small changes to the constants were required to truly minimize the SSE. It can be noted that the optimum value of Ea is much more accurately determined than that of Mo and Do.

#### 5.4.2.9 Comparison of the final model with the experimental data



**Figure 5.15: Fitting the Theoretical oil extraction model to experimental data using best-fit D values**

The final model was determined from the refining of Do, Mo and Ea to calculate the best coefficient from the Equation 5.13.



**Figure 5.16:** Fitting the Theoretical oil extraction model calculated to experimental data using D values calculated from refined Do and Ea constants

Graphical results for the plots of leaf oil mass concentration variation with time are shown in Figures 5.15 and 5.16 as derived from detailed calculations shown in Appendix C.

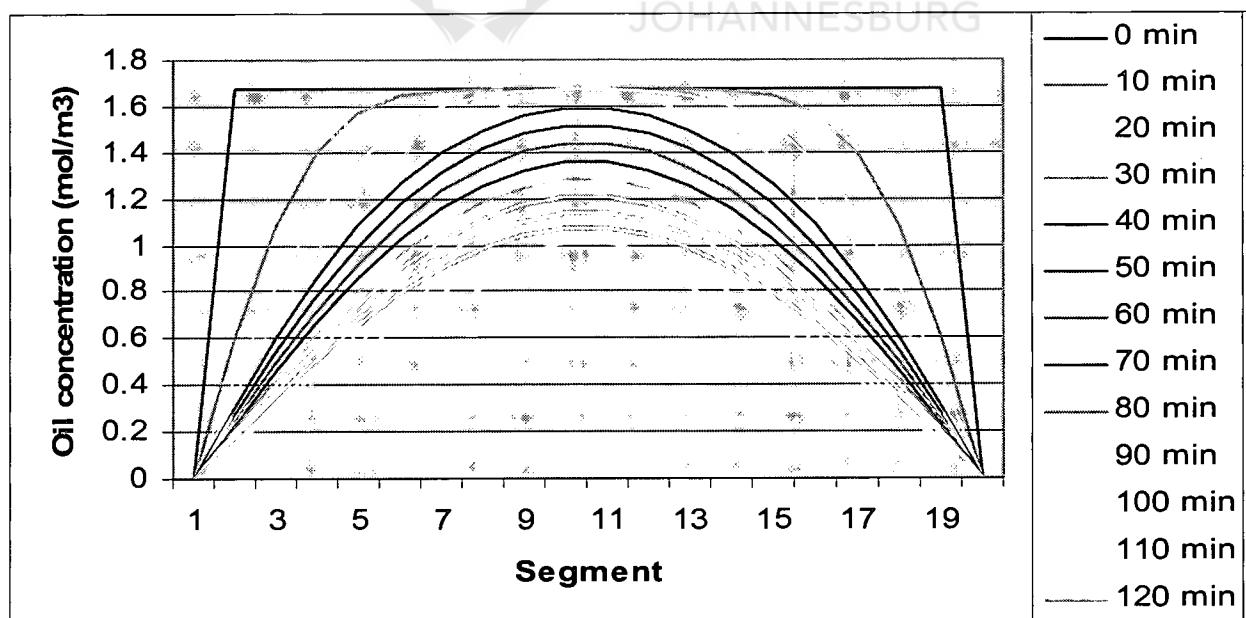
**Table 5.3 The literature values**

Essential oil	MW (g/mol)	Ea (kJ)	D ( $10^{-14} \text{ m}^2/\text{s}$ )	$D_o$ ( $10^8 \text{ m}^2/\text{s}$ )	T (K)
Orange oil	126.000	138.86	201.00	1.571334	363
Cinnamaladehyde	132.160	147.94	155.00	1.603562	373
Ginger oil	136.230	148.32	125.00	0.9731917	383
Citral	152.240	143.71	160.00	0.9403826	353
Cineole	154.249	138.17	161.00	2100049	343
Linalool	154.250	146.12	151.00	1312852	363
Lavender oil	154.250	139.44	105.00	1.3948380	373
Eucalyptus oil	154.299	131.19	187.00	1.1720162*	373
	154.299	135.90 <sup>+</sup>	161.00 <sup>+</sup>	1.0400000 <sup>+</sup>	373
Linalyl acetate	196.290	140.44	155.00	1.4892670	343
Cinnamol oil	288.000	151.05	140.00	1.0095940	323

\*Calculated from Equation 5.13

<sup>+</sup>Values from this work calculated for 373K for D.

Here it can be seen that the values obtained in this study are within the ballpark of literature values.



**Figure 5.17: Oil profiles in leaf cross-section during leaching using final model at 90°C**

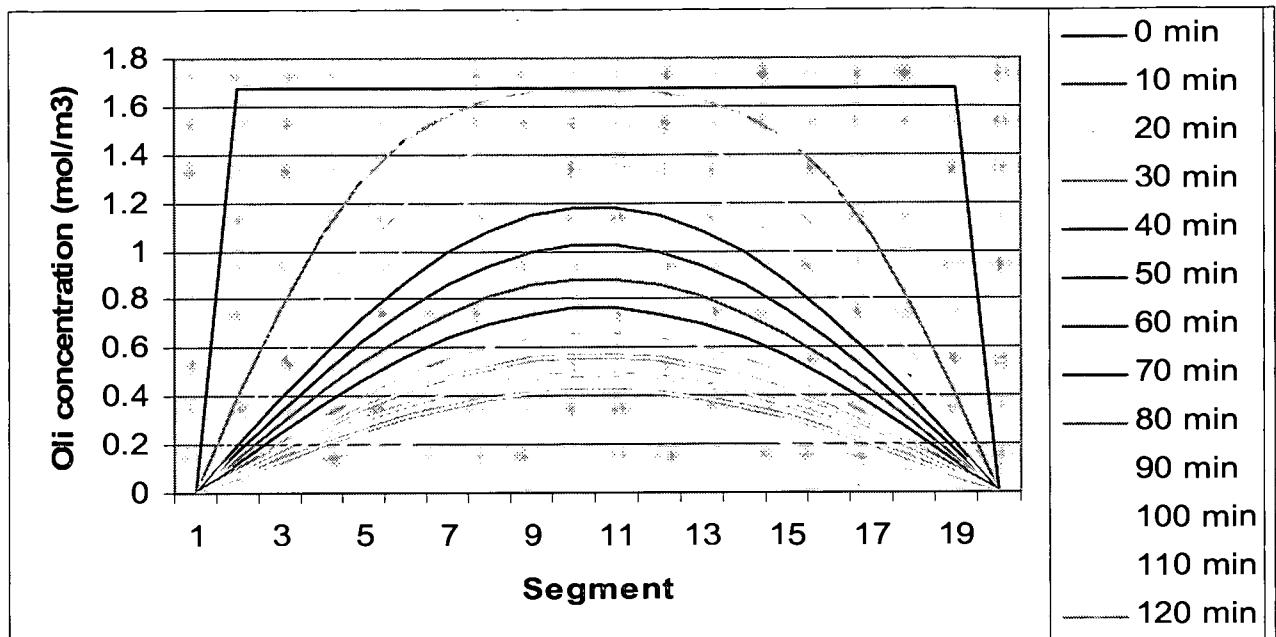


Figure 5.18 Oil profiles in leaf cross-section during leaching using final model at  $97^\circ\text{C}$

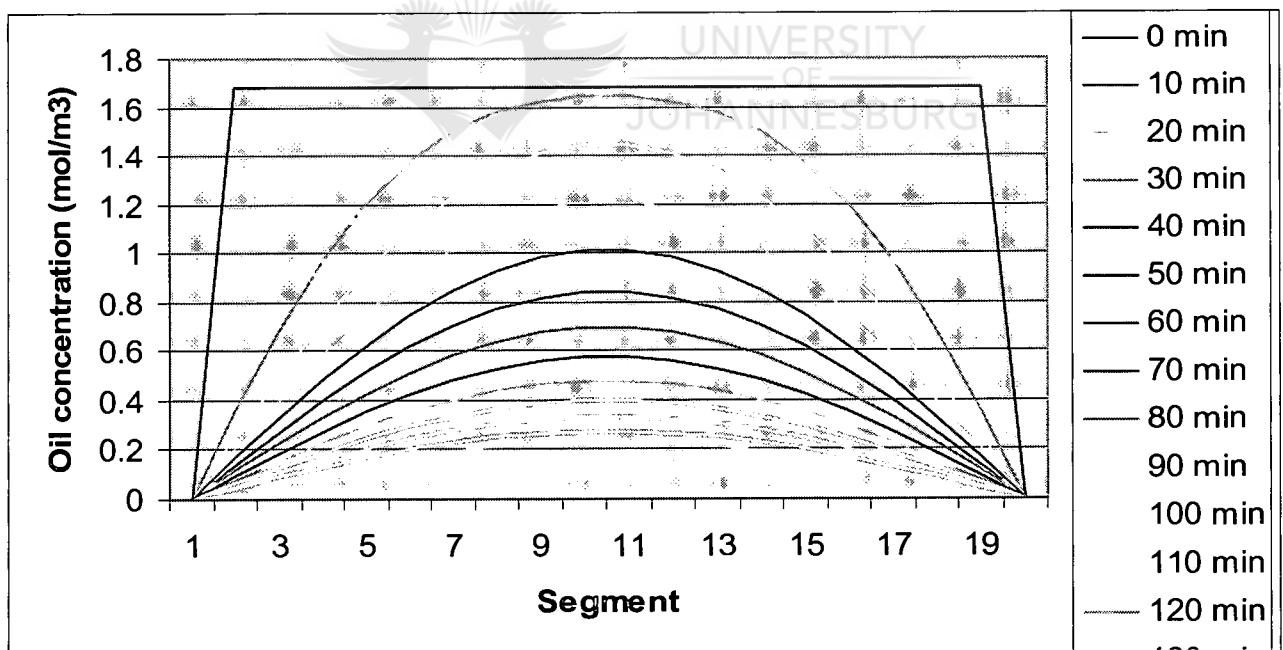


Figure 5.19: Oil profiles in leaf cross-section during leaching using final model at  $99^\circ\text{C}$

The Figures 5.17 -5.19 show the oil profiles in cross-section during leaching using the final model at the different temperatures used. Comparison of the figures indicates that oil is depleted more at the higher temperature.

The numerical method for the model was used to illustrate the change in concentration profile in leaf with various times during the extraction according to the Equation 5.12.

## 5.5 Concluding remarks

The Eucalyptus oil yields and quality obtained by the operating parameters subjected to the leaves have been presented and discussed here. The pressure at 150 kPa and temperature at 99°C were found to be the optimum operating pressure and temperature respectively. The yield was calculated from the relation between the essential oil mass extracted and the raw material mass used in the extraction. The volatiles released from the biomaterials, Eucalyptus, were analyzed using Gas Chromatography and eight major components were found to be contained in this species.

A mathematical model based on diffusion of essential oil in leaves was developed. Using a numerical method, the best diffusion coefficient was established for different operating conditions by comparing the model concentration of oil remaining in the leaves with the experimental amount of oil recovered; hence minimizing the sum of squared errors.

The Arrhenius model was used to account for the effect of temperature. The values determined for the constants agree well with published literature. The resulting expression for the diffusion coefficient as a function of temperature can now be used to model the large scale extraction of the essential oils from Eucalyptus leaves.

## **Chapter 6**

### **Conclusions and Recommendations**

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#### **6.1 Achievement of objectives**

This study followed an inter-disciplinary approach to investigate the effects of different operating parameters such as pressure, temperature and extraction time on the yield and quality of the Eucalyptus oil extracts obtained using steam distillation.

The yield of the oil extracts was determined in the form of the overall mass of oil recovered from an initial mass of 20kg of Eucalyptus leaf material. It was shown that an increase in temperature increases the extraction rate.

The quality of the oil extracts was monitored over the entire 120 minutes of the extraction process using Gas Chromatography. The Eucalyptus oil extracts were found to consist of 8 major components.

A mathematical model based on diffusion of essential oil in leaves was set up. Using a numerical method the best diffusion coefficient was established for different operating conditions by comparing the model concentration of oil leached in the leaves with the experimental amount of oil recovered; hence minimizing the sum of squared errors.

The model parameter evaluation, attained in this work to laboratorial scale, will be useful during the scale-up of the extraction process and/or during industrial operation to evaluate the extraction time required to obtain a given yield, as confirmed in this study.

As far as the amount of oil extracted is concerned, the best experimental conditions in this study were 150kPa, 99°C and 120 minutes of extraction, where the yield was 0.000255g/g

## **6.2 Contribution of the study**

It was possible to verify the importance of the process parameters in the yield of the Eucalyptus essential oil by steam distillation by using a model to determine the concentration profiles at any time in the leaves during the extraction. A yield curve was obtained for selected variables in the extraction of oil from natural plant material during a two hours long process.

The experimental data for Eucalyptus essential oil yields was well correlated by the diffusional model. The best fit adjusted parameter of the model (Diffusion coefficient) was in agreement with available literature data in terms of order of magnitude, backing up the model used. The proposed model revealed that the assumption that all the oil was removed during the experimental was not correct, as the amount of oil leached depended on the temperature of the test. Hence it is contended that, the model parameter evaluation, attained in this work to laboratory scale, will be useful during the scale-up of the extraction process and/or during pilot or industrial operation to evaluate the extraction conditions required to obtain a given yield.

## **6.3 Critical evaluation of the study**

It would have been beneficial to have data from more than two seasons to compare and collate. In addition, preliminary investigation indicated that monthly variations in the oil composition could be expected. This was however, not fully investigated. One of the reasons is that the onset of growth is dependent on the onset of precipitation. Another short coming of the investigation is that the oil compounds were not identified. This can make a further contribution to the medicinal value of the plant and should form part of further research.

A purely theoretical mathematical model (not a numerical method) could also have been developed for the steam extraction of Eucalyptus leaves. This model would also be used to fit the experimental data and could be used to confirm the values obtained for the diffusion coefficient in this work.

## 6.4 Recommendations for further research

In future studies,

- ❖ For the development of the purely theoretical mathematical model, three main steps could be hypothesized for the movement of the essential oil from the inside of the oil containing cells of the eucalyptus leaves, through the boundary layer and finally being dispersed into the bulk steam environment and defined as follows:
  - Diffusion of the oil from inside of the cells, through the cell walls, to the outer surface of the leaf material.
  - Diffusion of the oil extracts across the surface leaf oil and bulk steam interface film layer.
  - Dissolution of the oil extract into the bulk steam environment before being carried over to the collection unit by mass transport of the steam.

The rates of each of the above three steps individually affect the mass transfer of the oil extracts from the leaves into the steam and hence the yield of the extracted oil.

- ❖ A development of an appropriate design for the steam extraction process plant using the diffusion coefficient obtained as a function of temperature.

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[http://juniper.orst.edu/oils\\_abs.htm](http://juniper.orst.edu/oils_abs.htm).[Accessed 19 April 2005].

## **Appendix A: Experimental run data analyses**

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### **A.1 Quantitative analysis results**

The quantitative data were collected while extracting the Eucalyptus essential oils by weighing the quantities using an electronic analytical balance as described in Chapter 4. The following data was collected for the samples run for the various temperatures.

**Table A.1: Mass oil extracted**

<b>Time (min)</b>	<b>90°C</b>	<b>97°C</b>	<b>99°C</b>
0	0	0	0
10	0.58	1.30	1.10
20	1.25	2.00	2.22
30	1.50	2.50	2.90
40	2.10	2.70	3.25
50	2.25	3.00	3.88
60	2.40	3.50	4.25
70	2.63	3.83	4.33
80	2.75	4.00	4.52
90	2.82	4.02	4.56
100	2.95	4.06	4.62
110	3.00	4.10	4.68
120	3.25	4.50	5.10
130	3.25	4.50	5.10
140	3.25	4.50	5.10

**Table A. 2: Yield of mass oil extracted**

Time (min)	90oC	97oC	99oC
0	0	0	0
10	0.000029	0.000065	0.000055
20	6.25E-05	0.00010	0.000111
30	0.000075	0.000125	0.000145
40	0.000105	0.000135	0.000163
50	0.000113	0.00015	0.000194
60	0.00012	0.000175	0.000213
70	0.000132	0.000192	0.000217
80	0.000138	0.00020	0.000226
90	0.000141	0.000201	0.000228
100	0.000148	0.000203	0.000231
110	0.000150	0.000205	0.000234
120	0.000163	0.000225	0.000255
130	0.000163	0.000225	0.000255
140	0.000163	0.000225	0.000255

## A.2 Qualitative analysis results

### A.2.1 Raw run data for qualitative analysis

As described in chapter 4 of the experimental procedure for the analyses using Gas Chromatography, the quality of the oil was reported in terms of the areas of the peaks under the chromatogram trace. Each of the samples collected during the extraction period were injected separately to identify the different quantities of the components contained in them.

Each component of the mixture reaches the detector at a different time and produces a signal at a characteristic time called a retention time. The area under a peak is related to the amount of that

component present in the mixture. The number of peaks correlates with the number of components in the samples. The area under each peak correlates with the relative amount of that component in the sample. Chromatograph data is presented as a graph of detector response (y-axis) against retention time (x-axis).

Eight different major peaks were detected overall in all the injected samples and the corresponding areas recorded accordingly. The areas of the minor peaks were ignored and hence the results were corrected and normalised to that of the eight major peaks in order to determine the corresponding fractions in each injected sample. The peaks were numbered 1 to 8 for each injected sample according to their different retention times in the column.

#### A.2.1.1 Peak GC area data for qualitative analysis of the oil at 100kPa Boiler pressure



Table A3: Oil extracted raw qualitative GC data at 100kPa

Retention	Area	Area %	Height
2.091	99.1248	0.07	31.811
2.238	391.5974	0.28	114.460
2.501	221.0669	0.16	43.459
2.7196	28104.4060	20.02	3126.662
2.996	26198.1104	18.66	2749.093
3.258	45786.0189	32.62	4711.004
3.621	20488.1842	14.60	3149.497
3.898	277.0968	0.20	58.722
4.051	4930.5872	3.51	1054.922

4.165	2451.3524	1.75	635.335
4.318	615.8463	0.44	95.145
4.498	1944.0547	1.38	544.857
4.796	22.2472	0.02	7.426
4.928	356.7849	0.25	59.765
5.113	38.4760	0.03	7.926
5.465	218.5447	0.16	39.977
5.591	202.9972	0.14	49.693
5.945	14.4198	0.01	5.776
7.443	19.4259	0.01	6.442
7.538	16.2221	0.01	5.584
7.710	29.4827	0.02	8.248
7.971	854.0049	0.61	215.136
8.220	37.3676	0.03	9.755
8.381	52.2812	0.04	12.964
8.630	31.4498	0.02	8.866
8.916	63.0360	0.04	13.854
9.240	1026.6307	0.73	258.544
9.301	515.6679	0.37	168.699
9.405	2694.9496	1.92	646.524
9.673	104.5533	0.07	32.630
10.150	107.4786	0.08	20.500
10.518	120.7536	0.09	32.769
10650	38.0558	0.03	9.103

10.903	170.4888	0.12	43.689
11.241	285.3247	0.20	62.824
11.330	55.9410	0.04	16.873
11.486	258.6812	0.18	73.254
11.815	18.7727	0.01	5.539
12.135	10.8264	0.01	2.884
12.358	19.8825	0.01	6.425
12.475	13.025	0.01	4.834
12.713	20.8920	0.01	7.808
13.348	33.3580	0.02	7.234
13.560	24.2445	0.02	6.400
13.793	50.4541	0.04	14.693
13.860	65.1815	0.05	18.166
14.283	35.8618	0.03	6.758
14.355	41.4200	0.03	7.238
14.605	137.8514	0.10	29.158
14.845	17.5594	0.01	6.238
15.140	20.4456	0.01	4.391
15.370	14.5923	0.01	2.631
15.526	205.5158	0.15	50.265
15.920	10.7306	0.01	3.147
16.113	153.1129	0.11	37.203
16.205	154.6364	0.11	36.354
16.318	326.4350	0.23	70.308

16.406	125.3391	0.09	27.246
16.680	12.2356	0.01	2.619
16.881	20.3786	0.01	4.362

### A.2.1.2 Peak GC area data for qualitative analysis of the oil at 200 kPa Boiler pressure.

Table A.4: Oil extracted raw qualitative GC data at 200kPa

Retention	Area	Area (100%)	Height
2.085	120.8482	0.08	42.465
2.228	459.9812	0.31	149.994
2.486	251.4584	0.17	53.416
2.776	27420.2071	18.33	3240.263
2.968	26183.5096	17.50	2818.963
3.238	47050.8092	31.45	4735.439
3.623	21451.2868	14.34	3128.712
3.921	286.2097	0.19	59.168
4.085	5189.7176	3.47	1047.098
4.205	2563.3858	1.71	630.392
4.368	630.1157	0.42	91.772
5.558	2011.3796	1.34	541.894
4.870	25.8069	0.02	8.000
5.011	381.9369	0.26	62.027

5.196	46.7458	0.33	8.804
5.565	226.0640	0.15	40.989
5.695	209.3955	0.14	50.546
6.088	40.0131	0.03	9.465
7.278	10.2725	0.01	2.983
7.801	78.5140	0.05	15.773
8.041	35.0956	0.02	6.096
8.246	1448.8124	0.97	354.024
8.483	61.4468	0.04	14.838
8.726	44.4956	0.03	11.646
8.890	63.4537	0.04	16.927
9.175	83.9531	0.06	15.510
9.531	1260.9371	0.84	228.914
9.688	8452.5883	5.65	1482.158
9.911	196.6734	0.13	66.445
10.366	153.2006	0.10	30.218
10.733	168.7775	0.11	47.581
10.863	73.3852	0.05	17.806
11.115	370.2544	0.25	99.147
11.446	463.6334	0.31	95.086
11.525	74.5268	0.05	25.401
11.685	403.9816	0.27	115.959
12.003	35.9084	0.02	10.987
12.126	27.1506	0.02	8.667

12.533	26.8359	0.02	8.575
12.653	12.7800	0.01	4.866
12.886	19.7382	0.01	7.244
13.516	21.6718	0.01	4.963
13.721	28.7056	0.02	6.694
13.961	88.5148	0.06	25.436
14.026	79.1026	0.05	22.814
14.448	36.1560	0.02	7.093
14.535	67.9509	0.05	12.632
14.776	115.4212	0.08	26.344
15.300	18.7066	0.01	4.153
15.530	16.9135	0.01	3.607
15.683	233.7350	0.16	56.255
16.266	143.0797	0.10	33.877
16.361	122.8029	0.08	29.615
16.473	353.4463	0.24	76.842
16.563	125.9160	0.08	29.288
16.835	14.3282	0.01	3.049
17.035	15.6098	0.01	3.366

#### A.2.1.3 Peak GC area data for qualitative analysis of the Hexane solvent

**Table A.5: Hexane solvent qualitative GC data**

Retention	Area	Area %	Height
1.238	191.4281	0.14	226.146
1.261	826.1676	0.61	817.104
1.298	78311.2761	58.24	5001.865
1.638	34653.6963	25.77	2890.196
1.963	12820.5777	6.53	1243.714
2.208	4730.8350	3.52	629.200
2.458	207.4902	0.15	38.518
2.613	1019.0630	0.76	117.443
2.768	277.9468	0.21	34.775
2.943	77.2643	0.06	14.736
3.163	509.9028	0.38	50.824
3.343	447.3148	0.33	48.671
3.555	33.6208	0.03	5.093
3.868	128.7202	0.10	9.070
6.386	54.1272	0.04	8.420
6.788	48.5254	0.04	9.255
7.195	18.7485	0.01	3.338
7.391	16.3536	0.01	3.707
7.753	84.1537	0.06	17.272
8.275	12.7074	0.01	2.246

## **Appendix B: Modelling results and analyses**

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### **B.1. Fitting the Theoretical oil extraction model using the best fit D value**

#### **B.1.1 Fitting the Theoretical oil extraction model using the best fit D value to 90°C experimental data**

**Table B.1: Fitting the Theoretical oil extraction model using the best fit D value to 90°C experimental data**

Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	0.58	0.77	0.04
20.00	1.25	1.19	0.00
30.00	1.50	1.51	0.00
40.00	2.10	1.79	0.10
50.00	2.25	2.03	0.05
60.00	2.40	2.25	0.02
70.00	2.63	2.45	0.03
80.00	2.75	2.63	0.01
90.00	2.82	2.81	0.00
100.00	2.95	2.97	0.00
110.00	3.00	3.12	0.01
120.00	3.25	3.26	0.00
130.00	3.25	3.39	0.02
140.00	3.25	3.52	0.07
SUM (SSE) =			<b>0.36</b>

**Table B.2: Fitting the Theoretical oil extraction model calculated D values using Equation 5.17 to 90°C experimental data**

Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	0.58	0.76	0.03
20.00	1.25	1.17	0.01
30.00	1.50	1.49	0.00
40.00	2.10	1.77	0.11
50.00	2.25	2.01	0.06
60.00	2.40	2.22	0.03
70.00	2.63	2.42	0.04
80.00	2.75	2.61	0.02
90.00	2.82	2.78	0.00
100.00	2.95	2.94	0.00
110.00	3.00	3.09	0.01
120.00	3.25	3.23	0.00
130.00	3.25	3.36	0.01
140.00	3.25	3.48	0.05
SUM (SSE) =			0.38

**B.1.2 Fitting the Theoretical oil extraction model using the best fit D value to 97°C experimental data**

**Table B.3: Fitting the Theoretical oil extraction model using the best fit D value to 97°C experimental data**

Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	1.30	1.24	0.00
20.00	2.00	1.86	0.02
30.00	2.50	2.33	0.03
40.00	2.70	2.73	0.00
50.00	3.00	3.07	0.01
60.00	3.50	3.37	0.02
70.00	3.83	3.63	0.04
80.00	4.00	3.85	0.02

90.00	4.02	4.05	0.00
100.00	4.06	4.22	0.03
110.00	4.10	4.37	0.07
120.00	4.50	4.50	0.00
130.00	4.50	4.62	0.01
140.00	4.50	4.72	0.05
<b>SUM (SSE) =</b>			<b>0.30</b>

**Table B.4: Fitting the Theoretical oil extraction model calculated D values using Equation 5.17 to 97°C experimental data**

Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	1.30	1.30	0.00
20.00	2.00	1.94	0.00
30.00	2.50	2.43	0.00
40.00	2.70	2.84	0.02
50.00	3.00	3.19	0.04
60.00	3.50	3.49	0.00
70.00	3.83	3.75	0.01
80.00	4.00	3.98	0.00
90.00	4.02	4.17	0.02
100.00	4.06	4.34	0.08
110.00	4.10	4.48	0.15
120.00	4.50	4.61	0.01
130.00	4.50	4.72	0.05
140.00	4.50	4.81	0.10
<b>SUM (SSE) =</b>			<b>0.48</b>

**B.1.3 Fitting the Theoretical oil extraction model using the best fit D value to 99°C experimental data**

**Table B.5: Fitting the Theoretical oil extraction model using the best fit D value to 99°C experimental data**

Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	1.10	1.55	0.20
20.00	2.22	2.29	0.01
30.00	2.90	2.86	0.00
40.00	3.25	3.32	0.00
50.00	3.88	3.69	0.03
60.00	4.25	4.00	0.06
70.00	4.33	4.25	0.01
80.00	4.52	4.46	0.00
90.00	4.56	4.63	0.00
100.00	4.62	4.77	0.02
110.00	4.68	4.88	0.04
120.00	5.10	4.97	0.02
130.00	5.10	5.05	0.00
140.00	5.10	5.11	0.00
<b>SUM (SSE) =</b>			<b>0.40</b>

**Table B.6: Fitting the Theoretical oil extraction model calculated D values using Equation 5.17 to 99°C experimental data**

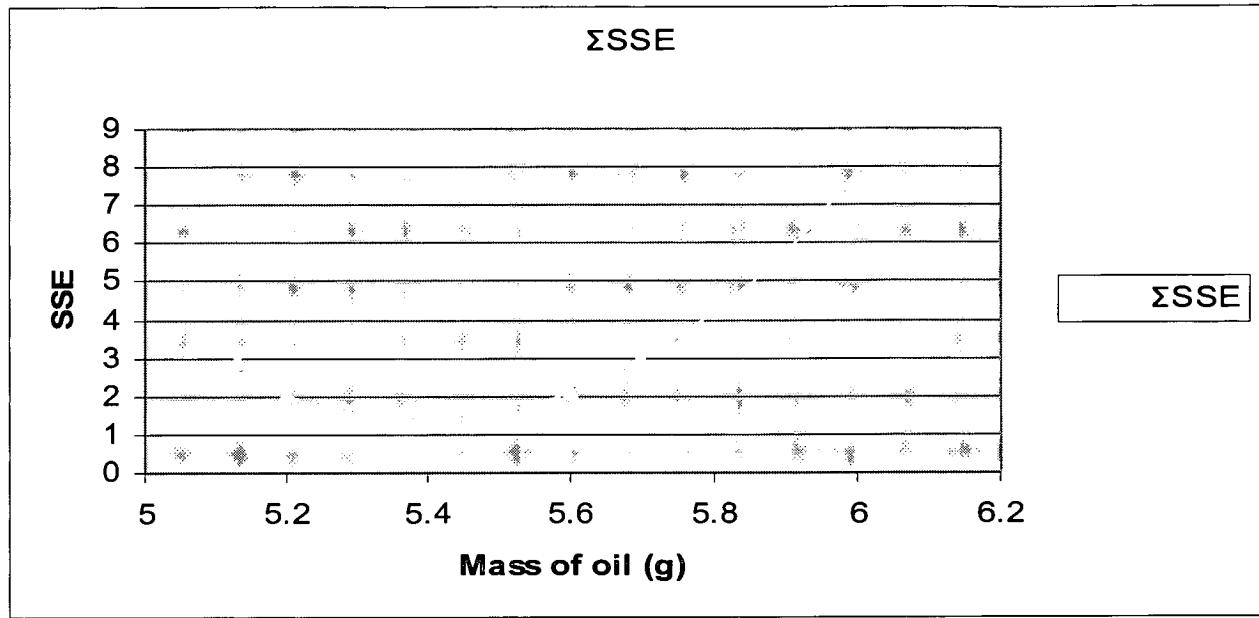
Time (min)	Mass Experimental	Mass Model	SSE
0.00	0.00	0.00	0.00
10.00	1.10	1.49	0.15
20.00	2.22	2.22	0.00
30.00	2.90	2.77	0.02
40.00	3.25	3.22	0.00
50.00	3.88	3.59	0.09
60.00	4.25	3.90	0.13
70.00	4.33	4.15	0.03

80.00	4.52	4.36	0.02	
90.00	4.56	4.54	0.00	
100.00	4.62	4.69	0.00	
110.00	4.68	4.81	0.02	
120.00	5.10	4.91	0.04	
130.00	5.10	4.99	0.01	
140.00	5.10	5.06	0.00	
<b>SUM (SSE) =</b>			<b>0.51</b>	

## B.2. Refining Mo, Do and Ea to ensure minimum SSE

Table B.7: Initial mass of oil leached versus SSE

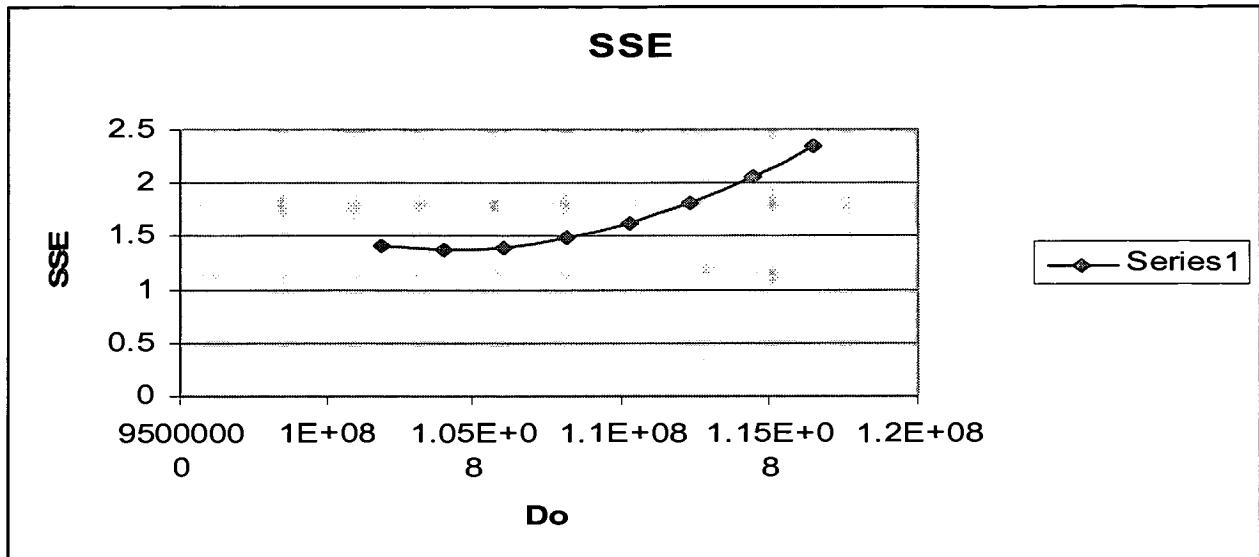
Mo (g)	$\Sigma$ SSE
5.13	2.92
5.20	2.05
5.30	1.54
5.40	1.37
5.50	1.56
5.60	2.10
5.70	2.98
5.80	4.22
5.90	5.81
6.00	7.75



**Figure B.1:** Initial mass of oil leached versus SSE

**Table B.8:** Maximum coefficient of diffusion versus SSE

Do (m <sup>2</sup> /s)	SSE
97760000	
99840000	
1.02E+08	1.41
1.04E+08	1.37
1.06E+08	1.40
1.08E+08	1.48
1.1E+08	1.62
1.12E+08	1.82
1.14E+08	2.06
1.16E+08	2.35



**Figure B.2: Maximum coefficient of diffusion versus SSE**

**Table B.8: Activation energy versus SSE**

Ea (kJ/mol)	SSE
100000	1.41
1.37	1.37
5.98	1.40
182	1.48
	1.62
	1.82
	2.06
	2.35

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**Table B.9: Optimization of Do, Mo and Ea versus SSE**

%	Do	Mo	Ea (kJ/mol)
94	1.69E+00	2.92	
96	1.514695	2.05	
98	1.41	1.54	100000
100	1.37	1.37	1.37
102	1.40	1.56	5.98
104	1.48	2.10	182
106	1.62	2.98	
108	1.82	4.22	
110	2.06	5.81	
112	2.35	7.75	

**Table B.10: Determination of the activation energy:  $\ln D$  versus  $1/T$**

X	Y	XY	$X^2$	$Y^2$
0.002687089	25.4	-0.06825	7.22044E-06	645.2111
0.002701607	25.8	-0.06963	7.29868E-06	664.3364
0.002753683	26.5	-0.07306	7.58177E-06	703.9687
<hr/>				
n	$\Sigma X$	$\Sigma Y$	$\Sigma(X^2)$	$\Sigma(Y^2)$
3	0.008142379	77.70814	2.21019E-05	2013.516
<hr/>				
$(\Sigma X)^2$	$(\Sigma Y)^2$	$\Sigma(XY)$		
6.62983E-05	6038.555	-0.21095		
<hr/>				
Slope, m = - 16340.89513	y-int, b = 18.44854116		r = -0.992653363	
$\ln D_0 = b = 18.44854$			$D_0 = 102825214.7$	
Ea/R = -16340.89513			Ea = 135.8582021	
<hr/>				
1/T		ln D		D
0.002687		-25.4010		9.30E-12
0.002702		-25.7747		6.40E-12
0.002754		-26.5324		3.00E-12

### B.3 Final model with experimental data

**Table B.11: Fitting the Theoretical oil extraction model to experimental data using best-fit D values**

<b>Mo</b>	5.4 g
<b>Do</b>	9.73E-09 m <sup>2</sup> /s
<b>Ea</b>	135.8582 kJ/mol
<b>Mm</b>	154.299g
<b>SSE</b>	1.07E+00

	<b>Mass Exp</b>	<b>Mass Mod.</b>	<b>Mass Exp</b>	<b>Mass Mod.</b>	<b>Mass Exp</b>	<b>Mass Mod.</b>
	T/ °C	D	T/ °C	D	T/ °C	D
<b>Time (min)</b>	90	1.02E-08	97.00	1.01642E-08	99	1.01618E-08
0	0	0	0.00	0.0000000000	0	0
10	0.58	0.77	1.30	1.2300000000	1.10	1.550824566
20	1.25	1.19	2.00	1.8400000000	2.22	2.297950608
30	1.50	1.51	2.50	2.3200000000	2.90	2.865945473
40	2.10	1.79	2.70	2.7100000000	3.25	3.324458961
50	2.25	2.03	3.00	3.0500000000	3.88	3.699127853
60	2.40	2.25	3.50	3.3500000000	4.25	4.006021636
70	2.63	2.45	3.83	3.6100000000	4.33	4.257518945
80	2.75	2.63	4.00	3.8400000000	4.52	4.463638249
90	2.82	2.81	4.02	4.0300000000	4.56	4.632570213
100	2.95	2.97	4.06	4.2100000000	4.62	4.771024537
110	3.00	3.12	4.10	4.3600000000	4.68	4.884499883
120	3.25	3.26	4.50	4.4900000000	5.10	4.977502801
130	3.25	3.39	4.50	4.6000000000	5.10	5.053726783
140	3.25	3.52	4.50	4.703132755	5.10	5.116198969

**Table B.12: Fitting the Theoretical oil extraction model calculated D values using Equation**

**5.13**

<b>Mo</b>	5.4 g
<b>Do</b>	1.04E+08 m <sup>2</sup> /s
<b>Ea</b>	135.8582 kJ/mol
<b>Mm</b>	154.299 g
<b>SSE</b>	1.37E+00

	<b>Mass Exp</b>	<b>Mass Mod.</b>	<b>Mass Exp</b>	<b>Mass Mod.</b>	<b>Mass Exp</b>	<b>Mass Mod.</b>
	<b>T/ °C</b>	<b>D</b>	<b>T/ °C</b>	<b>D</b>	<b>T/ °C</b>	<b>D</b>
<b>Time (min)</b>	90	2.94E-12	97.00	6.89403E-12	99	8.74062E-12
0	0	0	0.00	0.0000000000	0	0
10	0.58	0.77	1.30	1.2300000000	1.10	1.550824566
20	1.25	1.19	2.00	1.8400000000	2.22	2.297950608
30	1.50	1.51	2.50	2.3200000000	2.90	2.865945473
40	2.10	1.79	2.70	2.7100000000	3.25	3.324458961
50	2.25	2.03	3.00	3.0500000000	3.88	3.699127853
60	2.40	2.25	3.50	3.3500000000	4.25	4.006021636
70	2.63	2.45	3.83	3.6100000000	4.33	4.257518945
80	2.75	2.63	4.00	3.8400000000	4.52	4.463638249
90	2.82	2.81	4.02	4.0300000000	4.56	4.632570213
100	2.95	2.97	4.06	4.2100000000	4.62	4.771024537
110	3.00	3.12	4.10	4.3600000000	4.68	4.884499883
120	3.25	3.26	4.50	4.4900000000	5.10	4.977502801
130	3.25	3.39	4.50	4.6000000000	5.10	5.053726783
140	3.25	3.52	4.50	4.703132755	5.10	5.116198969

### B.3.1 Spreadsheet for the mass of oil leached at 90°C

<b>Segments</b>	18		
<b>Δx</b>	2.78E-05 m	<b>Total mass</b>	20 kg
<b>C<sub>0</sub></b>	1.68E+00 mol/m <sup>3</sup>	<b>Density</b>	960 kg/m <sup>3</sup>
<b>D</b>	2.94E-12 m <sup>2</sup> /sec	<b>Total Volume</b>	2.08E-02 m <sup>3</sup>
<b>Δt</b>	30 sec	<b>Thickness</b>	0.0005 m
<b>SSE:</b>	3.82E-01	<b>Total area</b>	41.66667 m <sup>2</sup>

#### B.3.1.1 Concentration (mol/m<sup>3</sup>) for the mass oil in the leaf at time Δt



Δt		Right edge	Leaf	Left edge





4110	137	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.17	1.03	0.9	0.68	0.46	0.2
4140	138	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.16	1.03	0.9	0.67	0.46	0.2
4170	139	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.16	1.03	0.9	0.67	0.46	0.2
4200	140	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.16	1.02	0.9	0.67	0.46	0.2
4230	141	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.15	1.02	0.9	0.67	0.46	0.2
4260	142	0	0.2	0.5	0.7	0.9	1	1.1	1.2	1.3	1.4	1.4	1.3	1.2	1.15	1.02	0.9	0.66	0.46	0.2
4290	143	0	0.2	0.5	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.14	1.01	0.8	0.66	0.45	0.2
4320	144	0	0.2	0.5	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.14	1.01	0.8	0.66	0.45	0.2
4350	145	0	0.2	0.5	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.14	1.01	0.8	0.66	0.45	0.2
4380	146	0	0.2	0.4	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.13	1	0.8	0.66	0.45	0.2
4410	147	0	0.2	0.4	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.13	1	0.8	0.65	0.45	0.2
4440	148	0	0.2	0.4	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.13	1	0.8	0.65	0.45	0.2
4470	149	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.12	0.99	0.8	0.65	0.44	0.2
4500	150	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.12	0.99	0.8	0.65	0.44	0.2
4530	151	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.12	0.99	0.8	0.64	0.44	0.2
4560	152	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.11	0.98	0.8	0.64	0.44	0.2
4590	153	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.11	0.98	0.8	0.64	0.44	0.2
4620	154	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.11	0.98	0.8	0.64	0.44	0.2
4650	155	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.1	0.97	0.8	0.63	0.43	0.2
4680	156	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.1	0.97	0.8	0.63	0.43	0.2
4710	157	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.1	0.97	0.8	0.63	0.43	0.2
4740	158	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.09	0.96	0.8	0.63	0.43	0.2
4770	159	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.2	1.09	0.96	0.8	0.63	0.43	0.2
4800	160	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.08	0.96	0.8	0.62	0.43	0.2
4830	161	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.08	0.95	0.8	0.62	0.43	0.2
4860	162	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.08	0.95	0.8	0.62	0.42	0.2
4890	163	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.07	0.95	0.8	0.62	0.42	0.2
4920	164	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.07	0.94	0.8	0.62	0.42	0.2
4950	165	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.07	0.94	0.8	0.61	0.42	0.2
4980	166	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.06	0.94	0.8	0.61	0.42	0.2
5010	167	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.06	0.94	0.8	0.61	0.42	0.2
5040	168	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.3	1.2	1.06	0.93	0.8	0.61	0.42	0.2
5070	169	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.2	1.2	1.2	1.2	1.05	0.93	0.8	0.6	0.41	0.2
5100	170	0	0.2	0.4	0.6	0.8	0.9	1.1	1.1	1.2	1.2	1.2	1.2	1.2	1.05	0.93	0.8	0.6	0.41	0.2
5130	171	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.2	1.05	0.92	0.8	0.6	0.41	0.2
5160	172	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.2	1.04	0.92	0.8	0.6	0.41	0.2
5190	173	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.2	1.04	0.92	0.8	0.6	0.41	0.2
5220	174	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.2	1.04	0.91	0.8	0.59	0.41	0.2
5250	175	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.1	1.03	0.91	0.8	0.59	0.41	0.2
5280	176	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.1	1.03	0.91	0.8	0.59	0.4	0.2

6360	212	0	0.2	0.4	0.5	0.7	0.8	0.92	1.01	1.1	1.09	1.1	1.1	1.01	0.9	0.8	0.7	0.53	0.4	0.18
6390	213	0	0.2	0.4	0.5	0.7	0.8	0.92	1	1.1	1.09	1.1	1.1	1	0.9	0.8	0.7	0.52	0.4	0.18
6420	214	0	0.2	0.4	0.5	0.7	0.8	0.92	1	1.1	1.09	1.1	1.1	1	0.9	0.8	0.7	0.52	0.4	0.18
6450	215	0	0.2	0.4	0.5	0.7	0.8	0.91	1	1.1	1.08	1.1	1.1	1	0.9	0.8	0.7	0.52	0.4	0.18
6480	216	0	0.2	0.4	0.5	0.7	0.8	0.91	0.99	1.1	1.08	1.1	1.1	0.99	0.9	0.8	0.7	0.52	0.4	0.18
6510	217	0	0.2	0.4	0.5	0.7	0.8	0.91	0.99	1	1.08	1.1	1	0.99	0.9	0.8	0.7	0.52	0.4	0.18
6540	218	0	0.2	0.4	0.5	0.7	0.8	0.9	0.99	1	1.07	1.1	1	0.99	0.9	0.8	0.7	0.52	0.4	0.18
6570	219	0	0.2	0.4	0.5	0.7	0.8	0.9	0.99	1	1.07	1.1	1	0.99	0.9	0.8	0.7	0.51	0.4	0.18
6600	220	0	0.2	0.3	0.5	0.7	0.8	0.9	0.98	1	1.07	1.1	1	0.98	0.9	0.8	0.7	0.51	0.3	0.18
6630	221	0	0.2	0.3	0.5	0.7	0.8	0.9	0.98	1	1.06	1.1	1	0.98	0.9	0.8	0.7	0.51	0.3	0.18
6660	222	0	0.2	0.3	0.5	0.7	0.8	0.89	0.98	1	1.06	1.1	1	0.98	0.9	0.8	0.7	0.51	0.3	0.18
6690	223	0	0.2	0.3	0.5	0.7	0.8	0.89	0.97	1	1.06	1.1	1	0.97	0.9	0.8	0.7	0.51	0.3	0.18
6720	224	0	0.2	0.3	0.5	0.7	0.8	0.89	0.97	1	1.05	1.1	1	0.97	0.9	0.8	0.7	0.51	0.3	0.18
6750	225	0	0.2	0.3	0.5	0.6	0.8	0.88	0.97	1	1.05	1.1	1	0.97	0.9	0.8	0.6	0.5	0.3	0.17
6780	226	0	0.2	0.3	0.5	0.6	0.8	0.88	0.96	1	1.05	1	1	0.96	0.9	0.8	0.6	0.5	0.3	0.17
6810	227	0	0.2	0.3	0.5	0.6	0.8	0.88	0.96	1	1.04	1	1	0.96	0.9	0.8	0.6	0.5	0.3	0.17
6840	228	0	0.2	0.3	0.5	0.6	0.8	0.88	0.96	1	1.04	1	1	0.96	0.9	0.8	0.6	0.5	0.3	0.17
6870	229	0	0.2	0.3	0.5	0.6	0.8	0.87	0.95	1	1.04	1	1	0.95	0.9	0.8	0.6	0.5	0.3	0.17
6900	230	0	0.2	0.3	0.5	0.6	0.8	0.87	0.95	1	1.04	1	1	0.95	0.9	0.8	0.6	0.5	0.3	0.17
6930	231	0	0.2	0.3	0.5	0.6	0.8	0.87	0.95	1	1.03	1	1	0.95	0.9	0.8	0.6	0.49	0.3	0.17
6960	232	0	0.2	0.3	0.5	0.6	0.8	0.87	0.95	1	1.03	1	1	0.95	0.9	0.8	0.6	0.49	0.3	0.17
6990	233	0	0.2	0.3	0.5	0.6	0.8	0.86	0.94	1	1.03	1	1	0.94	0.9	0.8	0.6	0.49	0.3	0.17
7020	234	0	0.2	0.3	0.5	0.6	0.8	0.86	0.94	1	1.02	1	1	0.94	0.9	0.8	0.6	0.49	0.3	0.17
7050	235	0	0.2	0.3	0.5	0.6	0.8	0.86	0.94	1	1.02	1	1	0.94	0.9	0.8	0.6	0.49	0.3	0.17
7080	236	0	0.2	0.3	0.5	0.6	0.8	0.85	0.93	1	1.02	1	1	0.93	0.9	0.8	0.6	0.49	0.3	0.17
7110	237	0	0.2	0.3	0.5	0.6	0.7	0.85	0.93	1	1.01	1	1	0.93	0.9	0.7	0.6	0.49	0.3	0.17
7140	238	0	0.2	0.3	0.5	0.6	0.7	0.85	0.93	1	1.01	1	1	0.93	0.8	0.7	0.6	0.48	0.3	0.17
7170	239	0	0.2	0.3	0.5	0.6	0.7	0.85	0.93	1	1.01	1	1	0.93	0.8	0.7	0.6	0.48	0.3	0.17
7200	240	0	0.2	0.3	0.5	0.6	0.7	0.84	0.92	1	1	1	1	0.92	0.8	0.7	0.6	0.48	0.3	0.17
7230	241	0	0.2	0.3	0.5	0.6	0.7	0.84	0.92	1	1	1	1	0.92	0.8	0.7	0.6	0.48	0.3	0.17
7260	242	0	0.2	0.3	0.5	0.6	0.7	0.84	0.92	1	1	1	1	0.92	0.8	0.7	0.6	0.48	0.3	0.17
7290	243	0	0.2	0.3	0.5	0.6	0.7	0.84	0.91	1	0.99	1	1	0.91	0.8	0.7	0.6	0.48	0.3	0.16
7320	244	0	0.2	0.3	0.5	0.6	0.7	0.83	0.91	1	0.99	1	1	0.91	0.8	0.7	0.6	0.47	0.3	0.16
7350	245	0	0.2	0.3	0.5	0.6	0.7	0.83	0.91	1	0.99	1	1	0.91	0.8	0.7	0.6	0.47	0.3	0.16
7380	246	0	0.2	0.3	0.5	0.6	0.7	0.83	0.91	1	0.98	1	1	0.91	0.8	0.7	0.6	0.47	0.3	0.16
7410	247	0	0.2	0.3	0.5	0.6	0.7	0.83	0.9	1	0.98	1	1	0.9	0.8	0.7	0.6	0.47	0.3	0.16
7440	248	0	0.2	0.3	0.5	0.6	0.7	0.82	0.9	1	0.98	1	1	0.9	0.8	0.7	0.6	0.47	0.3	0.16
7470	249	0	0.2	0.3	0.5	0.6	0.7	0.82	0.9	0.9	0.98	1	0.9	0.9	0.8	0.7	0.6	0.47	0.3	0.16
7500	250	0	0.2	0.3	0.5	0.6	0.7	0.82	0.89	0.9	0.97	1	0.9	0.89	0.8	0.7	0.6	0.47	0.3	0.16

7530	251	0	0.2	0.3	0.5	0.6	0.7	0.8	0.89	0.9	0.97	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7560	252	0	0.2	0.3	0.5	0.6	0.7	0.8	0.89	0.9	0.97	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7590	253	0	0.2	0.3	0.5	0.6	0.7	0.8	0.89	0.9	0.96	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7620	254	0	0.2	0.3	0.5	0.6	0.7	0.8	0.88	0.9	0.96	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7650	255	0	0.2	0.3	0.5	0.6	0.7	0.8	0.88	0.9	0.96	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7680	256	0	0.2	0.3	0.5	0.6	0.7	0.8	0.88	0.9	0.95	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7710	257	0	0.2	0.3	0.5	0.6	0.7	0.8	0.87	0.9	0.95	1	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7740	258	0	0.2	0.3	0.5	0.6	0.7	0.8	0.87	0.9	0.95	0.9	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7770	259	0	0.2	0.3	0.5	0.6	0.7	0.8	0.87	0.9	0.95	0.9	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7800	260	0	0.2	0.3	0.5	0.6	0.7	0.8	0.87	0.9	0.94	0.9	0.9	0.9	0.8	0.7	0.6	0.5	0.3	0.2	0
7830	261	0	0.2	0.3	0.4	0.6	0.7	0.8	0.86	0.9	0.94	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
7860	262	0	0.2	0.3	0.4	0.6	0.7	0.8	0.86	0.9	0.94	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
7890	263	0	0.2	0.3	0.4	0.6	0.7	0.8	0.86	0.9	0.93	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
7920	264	0	0.2	0.3	0.4	0.6	0.7	0.8	0.86	0.9	0.93	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
7950	265	0	0.2	0.3	0.4	0.6	0.7	0.8	0.85	0.9	0.93	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
7980	266	0	0.2	0.3	0.4	0.6	0.7	0.8	0.85	0.9	0.93	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0
8010	267	0	0.2	0.3	0.4	0.6	0.7	0.8	0.85	0.9	0.92	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8040	268	0	0.2	0.3	0.4	0.6	0.7	0.8	0.85	0.9	0.92	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8070	269	0	0.2	0.3	0.4	0.6	0.7	0.8	0.84	0.9	0.92	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8100	270	0	0.2	0.3	0.4	0.6	0.7	0.8	0.84	0.9	0.91	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8130	271	0	0.2	0.3	0.4	0.6	0.7	0.8	0.84	0.9	0.91	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8160	272	0	0.2	0.3	0.4	0.6	0.7	0.8	0.83	0.9	0.91	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.2	0
8190	273	0	0.1	0.3	0.4	0.6	0.7	0.8	0.83	0.9	0.91	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.1	0
8220	274	0	0.1	0.3	0.4	0.6	0.7	0.8	0.83	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.1	0
8250	275	0	0.1	0.3	0.4	0.6	0.7	0.8	0.83	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.1	0
8280	276	0	0.1	0.3	0.4	0.6	0.7	0.8	0.82	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.1	0
8310	277	0	0.1	0.3	0.4	0.6	0.7	0.8	0.82	0.9	0.89	0.9	0.9	0.8	0.8	0.7	0.6	0.4	0.3	0.1	0
8340	278	0	0.1	0.3	0.4	0.5	0.7	0.7	0.82	0.9	0.89	0.9	0.9	0.8	0.7	0.7	0.5	0.4	0.3	0.1	0
8370	279	0	0.1	0.3	0.4	0.5	0.7	0.7	0.82	0.9	0.89	0.9	0.9	0.8	0.7	0.7	0.5	0.4	0.3	0.1	0
8400	280	0	0.1	0.3	0.4	0.5	0.7	0.7	0.81	0.9	0.89	0.9	0.9	0.8	0.7	0.7	0.5	0.4	0.3	0.1	0

### B.3.1.2 Mass of oil leached

$\Delta t$		Right edge	Leaf	Left edge











227	0	0.031	0.061	0.089	0.115	0.138	0.157	0.172	0.18	0.19	0.19	0.182	0.172	0.157	0.14	0.115	0.09
228	0	0.031	0.061	0.089	0.115	0.138	0.156	0.171	0.18	0.19	0.19	0.181	0.171	0.156	0.14	0.115	0.09
229	0	0.031	0.061	0.089	0.115	0.137	0.156	0.171	0.18	0.19	0.19	0.18	0.171	0.156	0.14	0.115	0.09
230	0	0.031	0.06	0.089	0.114	0.137	0.155	0.17	0.18	0.18	0.18	0.18	0.17	0.155	0.14	0.114	0.09
231	0	0.031	0.06	0.088	0.114	0.136	0.155	0.169	0.18	0.18	0.18	0.179	0.169	0.155	0.14	0.114	0.09
232	0	0.03	0.06	0.088	0.114	0.136	0.155	0.169	0.18	0.18	0.18	0.179	0.169	0.155	0.14	0.114	0.09
233	0	0.03	0.06	0.088	0.113	0.135	0.154	0.168	0.18	0.18	0.18	0.178	0.168	0.154	0.14	0.113	0.09
234	0	0.03	0.06	0.087	0.113	0.135	0.154	0.168	0.18	0.18	0.18	0.178	0.168	0.154	0.14	0.113	0.09
235	0	0.03	0.06	0.087	0.112	0.135	0.153	0.167	0.18	0.18	0.18	0.177	0.167	0.153	0.13	0.112	0.09
236	0	0.03	0.059	0.087	0.112	0.134	0.153	0.167	0.18	0.18	0.18	0.177	0.167	0.153	0.13	0.112	0.09
237	0	0.03	0.059	0.087	0.112	0.134	0.152	0.166	0.18	0.18	0.18	0.176	0.166	0.152	0.13	0.112	0.09
238	0	0.03	0.059	0.086	0.111	0.133	0.152	0.166	0.18	0.18	0.18	0.175	0.166	0.152	0.13	0.111	0.09
239	0	0.03	0.059	0.086	0.111	0.133	0.151	0.165	0.17	0.18	0.18	0.175	0.165	0.151	0.13	0.111	0.09
240	0	0.03	0.059	0.086	0.111	0.133	0.151	0.165	0.17	0.18	0.18	0.174	0.165	0.151	0.13	0.111	0.09
241	0	0.03	0.058	0.086	0.11	0.132	0.15	0.164	0.17	0.18	0.18	0.174	0.164	0.15	0.13	0.11	0.09
242	0	0.03	0.058	0.085	0.11	0.132	0.15	0.164	0.17	0.18	0.18	0.173	0.164	0.15	0.13	0.11	0.09
243	0	0.029	0.058	0.085	0.11	0.131	0.149	0.163	0.17	0.18	0.18	0.173	0.163	0.149	0.13	0.11	0.08
244	0	0.029	0.058	0.085	0.109	0.131	0.149	0.163	0.17	0.18	0.18	0.172	0.163	0.149	0.13	0.109	0.08
245	0	0.029	0.058	0.084	0.109	0.13	0.148	0.162	0.17	0.18	0.18	0.172	0.162	0.148	0.13	0.109	0.08
246	0	0.029	0.057	0.084	0.109	0.13	0.148	0.162	0.17	0.18	0.18	0.171	0.162	0.148	0.13	0.109	0.08
247	0	0.029	0.057	0.084	0.108	0.13	0.147	0.161	0.17	0.18	0.18	0.171	0.161	0.147	0.13	0.108	0.08
248	0	0.029	0.057	0.084	0.108	0.129	0.147	0.161	0.17	0.17	0.17	0.17	0.161	0.147	0.13	0.108	0.08
249	0	0.029	0.057	0.083	0.108	0.129	0.147	0.16	0.17	0.17	0.17	0.17	0.16	0.147	0.13	0.108	0.08
250	0	0.029	0.057	0.083	0.107	0.128	0.146	0.16	0.17	0.17	0.17	0.169	0.16	0.146	0.13	0.107	0.08
251	0	0.029	0.057	0.083	0.107	0.128	0.146	0.159	0.17	0.17	0.17	0.168	0.159	0.146	0.13	0.107	0.08
252	0	0.029	0.056	0.083	0.107	0.128	0.145	0.159	0.17	0.17	0.17	0.168	0.159	0.145	0.13	0.107	0.08
253	0	0.028	0.056	0.082	0.106	0.127	0.145	0.158	0.17	0.17	0.17	0.167	0.158	0.145	0.13	0.106	0.08
254	0	0.028	0.056	0.082	0.106	0.127	0.144	0.158	0.17	0.17	0.17	0.167	0.158	0.144	0.13	0.106	0.08
255	0	0.028	0.056	0.082	0.106	0.126	0.144	0.157	0.17	0.17	0.17	0.166	0.157	0.144	0.13	0.106	0.08
256	0	0.028	0.056	0.082	0.105	0.126	0.143	0.157	0.17	0.17	0.17	0.166	0.157	0.143	0.13	0.105	0.08
257	0	0.028	0.055	0.081	0.105	0.126	0.143	0.156	0.17	0.17	0.17	0.165	0.156	0.143	0.13	0.105	0.08
258	0	0.028	0.055	0.081	0.105	0.125	0.142	0.156	0.16	0.17	0.17	0.165	0.156	0.142	0.13	0.105	0.08
259	0	0.028	0.055	0.081	0.104	0.125	0.142	0.155	0.16	0.17	0.17	0.164	0.155	0.142	0.12	0.104	0.08
260	0	0.028	0.055	0.081	0.104	0.124	0.142	0.155	0.16	0.17	0.17	0.164	0.155	0.142	0.12	0.104	0.08
261	0	0.028	0.055	0.08	0.104	0.124	0.141	0.154	0.16	0.17	0.17	0.163	0.154	0.141	0.12	0.104	0.08
262	0	0.028	0.055	0.08	0.103	0.124	0.141	0.154	0.16	0.17	0.17	0.163	0.154	0.141	0.12	0.103	0.08
263	0	0.028	0.054	0.08	0.103	0.123	0.14	0.153	0.16	0.17	0.17	0.162	0.153	0.14	0.12	0.103	0.08
264	0	0.028	0.054	0.08	0.103	0.123	0.14	0.153	0.16	0.17	0.17	0.162	0.153	0.14	0.12	0.103	0.08
265	0	0.027	0.054	0.079	0.102	0.123	0.139	0.152	0.16	0.17	0.17	0.161	0.152	0.139	0.12	0.102	0.08
266	0	0.027	0.054	0.079	0.102	0.122	0.139	0.152	0.16	0.17	0.17	0.161	0.152	0.139	0.12	0.102	0.08
267	0	0.027	0.054	0.079	0.102	0.122	0.138	0.151	0.16	0.16	0.16	0.151	0.151	0.138	0.12	0.102	0.08
268	0	0.027	0.054	0.079	0.101	0.121	0.138	0.151	0.16	0.16	0.16	0.151	0.151	0.138	0.12	0.101	0.08
269	0	0.027	0.053	0.078	0.101	0.121	0.138	0.151	0.16	0.16	0.16	0.159	0.151	0.138	0.12	0.101	0.08

040	268	0	0.027	0.05	0.079	0.101	0.12	0.138	0.151	0.16	0.164	0.164	0.16	0.151	0.14	0.121	0.101	0.08
070	269	0	0.027	0.05	0.078	0.101	0.12	0.138	0.151	0.159	0.164	0.164	0.16	0.151	0.14	0.121	0.101	0.08
100	270	0	0.027	0.05	0.078	0.101	0.12	0.137	0.15	0.159	0.163	0.163	0.16	0.15	0.14	0.121	0.101	0.08
130	271	0	0.027	0.05	0.078	0.1	0.12	0.137	0.15	0.158	0.163	0.163	0.16	0.15	0.14	0.12	0.1	0.08
160	272	0	0.027	0.05	0.078	0.1	0.12	0.136	0.149	0.158	0.162	0.162	0.16	0.149	0.14	0.12	0.1	0.08
190	273	0	0.027	0.05	0.077	0.1	0.12	0.136	0.149	0.157	0.162	0.162	0.16	0.149	0.14	0.119	0.1	0.08
220	274	0	0.027	0.05	0.077	0.099	0.12	0.135	0.148	0.157	0.161	0.161	0.16	0.148	0.14	0.119	0.099	0.08
250	275	0	0.027	0.05	0.077	0.099	0.12	0.135	0.148	0.156	0.161	0.161	0.16	0.148	0.14	0.119	0.099	0.08
280	276	0	0.026	0.05	0.077	0.099	0.12	0.135	0.147	0.156	0.16	0.16	0.16	0.147	0.13	0.118	0.099	0.08
310	277	0	0.026	0.05	0.076	0.099	0.12	0.134	0.147	0.155	0.16	0.16	0.16	0.147	0.13	0.118	0.099	0.08
340	278	0	0.026	0.05	0.076	0.098	0.12	0.134	0.146	0.155	0.159	0.159	0.15	0.146	0.13	0.118	0.098	0.08
370	279	0	0.026	0.05	0.076	0.098	0.12	0.133	0.146	0.154	0.159	0.159	0.15	0.146	0.13	0.117	0.098	0.08
400	280	0	0.026	0.05	0.076	0.098	0.12	0.133	0.145	0.154	0.158	0.158	0.15	0.145	0.13	0.117	0.098	0.08

### B.3.2 Spreadsheet for the mass of oil leached at 97°C

Segments	18		
$\Delta x$	2.78E-05 m		
$C_0$	1.68E+00 mol/m <sup>3</sup>		
D	6.89E-12 m <sup>2</sup> /sec		
$\Delta t$	30 sec		
SSE:	4.77E-01		
		Total mass	20 kg
		Density	960 kg/m <sup>3</sup>
		Total Volume	2.08E-02 m <sup>3</sup>
		Thickness	0.0005 m
		Total area	41.66667 m <sup>2</sup>

#### B.3.2.1 Concentration (mol/m<sup>3</sup>) for the mass oil in the leaf at time $\Delta t$

$\Delta t$		Right edge	Leaf	Left edge

0	0	0	1.7	1.7	1.7	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	0	
30	1	0	1.2	1.7	1.7	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.2	0
60	2	0	1	1.6	1.7	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1	0	
90	3	0	0.9	1.4	1.6	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.4	0.9	0	
120	4	0	0.8	1.4	1.6	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.4	0.8	0	
150	5	0	0.7	1.3	1.6	1.7	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.3	0.7	0		
180	6	0	0.7	1.2	1.5	1.6	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.2	0.7	0		
210	7	0	0.6	1.1	1.5	1.6	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.1	0.6	0		
240	8	0	0.6	1.1	1.4	1.6	1.7	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.4	1.1	0.6	0			
270	9	0	0.6	1	1.4	1.6	1.6	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.4	1	0.6	0				
300	10	0	0.5	1	1.3	1.5	1.6	1.7	1.68	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.3	1	0.5	0				
330	11	0	0.5	1	1.3	1.5	1.6	1.7	1.67	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.3	1	0.5	0					
360	12	0	0.5	0.9	1.3	1.5	1.6	1.6	1.67	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	0.9	0.5	0						
390	13	0	0.5	0.9	1.2	1.5	1.6	1.6	1.67	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.2	0.9	0.5	0						
420	14	0	0.5	0.9	1.2	1.4	1.6	1.6	1.66	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.4	1.2	0.9	0.5	0					
450	15	0	0.5	0.9	1.2	1.4	1.5	1.6	1.66	1.7	1.7	1.7	1.7	1.6	1.5	1.4	1.2	0.9	0.5	0						
480	16	0	0.4	0.8	1.2	1.4	1.5	1.6	1.65	1.7	1.7	1.7	1.7	1.6	1.5	1.4	1.2	0.8	0.4	0						
510	17	0	0.4	0.8	1.1	1.4	1.5	1.6	1.64	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.4	1.1	0.8	0.4	0					
540	18	0	0.4	0.8	1.1	1.3	1.5	1.6	1.64	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0					
570	19	0	0.4	0.8	1.1	1.3	1.5	1.6	1.63	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0					
600	20	0	0.4	0.8	1.1	1.3	1.5	1.6	1.62	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0					
630	21	0	0.4	0.7	1	1.3	1.4	1.6	1.62	1.6	1.7	1.7	1.6	1.6	1.6	1.4	1.3	1	0.7	0.4	0					
660	22	0	0.4	0.7	1	1.3	1.4	1.5	1.61	1.6	1.7	1.7	1.6	1.6	1.5	1.4	1.3	1	0.7	0.4	0					
690	23	0	0.4	0.7	1	1.2	1.4	1.5	1.6	1.6	1.7	1.7	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0					
720	24	0	0.4	0.7	1	1.2	1.4	1.5	1.59	1.6	1.7	1.7	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0					
750	25	0	0.4	0.7	1	1.2	1.4	1.5	1.58	1.6	1.6	1.6	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0					
780	26	0	0.4	0.7	1	1.2	1.4	1.5	1.57	1.6	1.6	1.6	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0					
810	27	0	0.3	0.7	0.9	1.2	1.4	1.5	1.56	1.6	1.6	1.6	1.6	1.6	1.5	1.4	1.2	0.9	0.7	0.3	0					
840	28	0	0.3	0.7	0.9	1.2	1.3	1.5	1.56	1.6	1.6	1.6	1.6	1.6	1.5	1.3	1.2	0.9	0.7	0.3	0					
870	29	0	0.3	0.6	0.9	1.2	1.3	1.5	1.55	1.6	1.6	1.6	1.6	1.6	1.5	1.3	1.2	0.9	0.6	0.3	0					
900	30	0	0.3	0.6	0.9	1.1	1.3	1.4	1.54	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
930	31	0	0.3	0.6	0.9	1.1	1.3	1.4	1.53	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
960	32	0	0.3	0.6	0.9	1.1	1.3	1.4	1.52	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
990	33	0	0.3	0.6	0.9	1.1	1.3	1.4	1.51	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
1020	34	0	0.3	0.6	0.9	1.1	1.3	1.4	1.5	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
1050	35	0	0.3	0.6	0.9	1.1	1.3	1.4	1.49	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0					
1080	36	0	0.3	0.6	0.8	1.1	1.2	1.4	1.48	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1.1	0.8	0.6	0.3	0					
1110	37	0	0.3	0.6	0.8	1.1	1.2	1.4	1.47	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1.1	0.8	0.6	0.3	0					
1140	38	0	0.3	0.6	0.8	1	1.2	1.4	1.46	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1	0.8	0.6	0.3	0					
1170	39	0	0.3	0.6	0.8	1	1.2	1.4	1.45	1.5	1.5	1.5	1.5	1.5	1.4	1.2	1	0.8	0.6	0.3	0					
1200	40	0	0.3	0.6	0.8	1	1.2	1.3	1.44	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.6	0.3	0					
1230	41	0	0.3	0.6	0.8	1	1.2	1.3	1.43	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.6	0.3	0					
1260	42	0	0.3	0.5	0.8	1	1.2	1.3	1.42	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0					
1290	43	0	0.3	0.5	0.8	1	1.2	1.3	1.41	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0					
1320	44	0	0.3	0.5	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0					
1350	45	0	0.3	0.5	0.8	1	1.1	1.3	1.39	1.5	1.5	1.5	1.5	1.4	1.3	1.1	1	0.8	0.5	0.3	0					
1380	46	0	0.3	0.5	0.8	1	1.1	1.3	1.38	1.5	1.5	1.5	1.5	1.4	1.3	1.1	1	0.8	0.5	0.3	0					









010	267	0	0	0.1	0.1	0.18	0.2	0.3	0.3	0.29	0.3	0.3	0.3	0.28	0.3	0.2	0.2	0.1	0.1	0	0
040	268	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.29	0.3	0.3	0.3	0.27	0.2	0.2	0.2	0.1	0.1	0	0
070	269	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.29	0.3	0.3	0.3	0.27	0.2	0.2	0.2	0.1	0.1	0	0
100	270	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.29	0.3	0.3	0.3	0.27	0.2	0.2	0.2	0.1	0.1	0	0
130	271	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.28	0.3	0.3	0.3	0.27	0.2	0.2	0.2	0.1	0.1	0	0
160	272	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.28	0.3	0.3	0.3	0.27	0.2	0.2	0.2	0.1	0.1	0	0
190	273	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.28	0.3	0.3	0.3	0.26	0.2	0.2	0.2	0.1	0.1	0	0
220	274	0	0	0.1	0.1	0.18	0.2	0.2	0.3	0.28	0.3	0.3	0.3	0.26	0.2	0.2	0.2	0.1	0.1	0	0
250	275	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.26	0.2	0.2	0.2	0.1	0.1	0	0
280	276	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.26	0.2	0.2	0.2	0.1	0.1	0	0
310	277	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.26	0.2	0.2	0.2	0.1	0.1	0	0
340	278	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.25	0.2	0.2	0.2	0.1	0.1	0	0
370	279	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.25	0.2	0.2	0.2	0.1	0.1	0	0
400	280	0	0	0.1	0.1	0.17	0.2	0.2	0.3	0.27	0.3	0.3	0.3	0.25	0.2	0.2	0.2	0.1	0.1	0	0

### B.3.2.2 Mass of oil leached

$\Delta t$		Right edge	Leaf OF JOHANNESBURG	Left edge

0	0	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	
1	0	0.22	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	
2	0	0.18	0.28	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.278	
3	0	0.16	0.26	0.294	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.258	
4	0	0.14	0.24	0.286	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.298	0.286	0.241
5	0	0.13	0.23	0.277	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.296	0.277	0.227
6	0	0.12	0.21	0.269	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.292	0.269	0.215	
7	0	0.11	0.2	0.261	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.287	0.261	0.204	
8	0	0.11	0.2	0.253	0.28	0.3	0.299	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.283	0.253	0.195	
9	0	0.1	0.19	0.245	0.28	0.29	0.298	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.278	0.245	0.187			
10	0	0.1	0.18	0.239	0.27	0.29	0.297	0.299	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.273	0.239	0.18			
11	0	0.09	0.17	0.232	0.27	0.29	0.296	0.299	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.269	0.232	0.174				
12	0	0.09	0.17	0.226	0.26	0.28	0.295	0.298	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.264	0.226	0.168				
13	0	0.09	0.16	0.221	0.26	0.28	0.293	0.298	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.259	0.221	0.163				
14	0	0.08	0.16	0.215	0.26	0.28	0.291	0.297	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.255	0.215	0.158				
15	0	0.08	0.15	0.211	0.25	0.28	0.289	0.296	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.251	0.211	0.153				
16	0	0.08	0.15	0.206	0.25	0.27	0.287	0.295	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.29	0.27	0.247	0.206	0.149			
17	0	0.08	0.15	0.202	0.24	0.27	0.285	0.294	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.29	0.27	0.243	0.202	0.146			
18	0	0.07	0.14	0.198	0.24	0.27	0.283	0.293	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.27	0.239	0.198	0.142			
19	0	0.07	0.14	0.194	0.24	0.26	0.281	0.291	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.26	0.235	0.194	0.139			
20	0	0.07	0.14	0.19	0.23	0.26	0.279	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.26	0.232	0.19	0.136			
21	0	0.07	0.13	0.187	0.23	0.26	0.277	0.289	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.26	0.228	0.187	0.133			
22	0	0.07	0.13	0.184	0.23	0.26	0.275	0.287	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.28	0.26	0.225	0.184	0.13			
23	0	0.07	0.13	0.181	0.22	0.25	0.273	0.286	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.27	0.25	0.222	0.181	0.128			
24	0	0.07	0.13	0.178	0.22	0.25	0.271	0.284	0.29	0.29	0.29	0.29	0.3	0.28	0.27	0.25	0.219	0.178	0.126					
25	0	0.06	0.12	0.175	0.22	0.25	0.269	0.283	0.29	0.29	0.29	0.29	0.3	0.28	0.27	0.25	0.216	0.175	0.123					
26	0	0.06	0.12	0.172	0.21	0.24	0.267	0.281	0.29	0.29	0.29	0.29	0.3	0.28	0.27	0.24	0.213	0.172	0.121					
27	0	0.06	0.12	0.17	0.21	0.24	0.265	0.279	0.29	0.29	0.29	0.29	0.3	0.28	0.26	0.24	0.211	0.17	0.119					
28	0	0.06	0.12	0.167	0.21	0.24	0.263	0.278	0.29	0.29	0.29	0.29	0.3	0.28	0.26	0.24	0.208	0.167	0.117					
29	0	0.06	0.12	0.165	0.21	0.24	0.261	0.276	0.29	0.29	0.29	0.29	0.3	0.28	0.26	0.24	0.206	0.165	0.115					
30	0	0.06	0.11	0.162	0.2	0.24	0.259	0.274	0.28	0.29	0.29	0.29	0.3	0.27	0.26	0.24	0.203	0.162	0.114					
31	0	0.06	0.11	0.16	0.2	0.23	0.257	0.273	0.28	0.29	0.29	0.29	0.3	0.27	0.26	0.23	0.201	0.16	0.112					
32	0	0.06	0.11	0.158	0.2	0.23	0.255	0.271	0.28	0.29	0.29	0.29	0.3	0.27	0.25	0.23	0.199	0.158	0.11					
33	0	0.06	0.11	0.156	0.2	0.23	0.253	0.269	0.28	0.28	0.28	0.28	0.3	0.27	0.25	0.23	0.196	0.156	0.109					
34	0	0.06	0.11	0.154	0.19	0.23	0.251	0.268	0.28	0.28	0.28	0.28	0.3	0.27	0.25	0.23	0.194	0.154	0.107					
35	0	0.05	0.11	0.152	0.19	0.22	0.249	0.266	0.28	0.28	0.28	0.28	0.3	0.27	0.25	0.22	0.192	0.152	0.106					
36	0	0.05	0.1	0.151	0.19	0.22	0.247	0.264	0.28	0.28	0.28	0.28	0.3	0.26	0.25	0.22	0.19	0.151	0.105					
37	0	0.05	0.1	0.149	0.19	0.22	0.245	0.263	0.27	0.28	0.28	0.28	0.3	0.26	0.24	0.22	0.188	0.149	0.103					
38	0	0.05	0.1	0.147	0.19	0.22	0.243	0.261	0.27	0.28	0.28	0.28	0.3	0.26	0.24	0.22	0.186	0.147	0.102					
39	0	0.05	0.1	0.145	0.18	0.22	0.241	0.259	0.27	0.28	0.28	0.28	0.3	0.26	0.24	0.22	0.184	0.145	0.101					
40	0	0.05	0.1	0.144	0.18	0.21	0.239	0.257	0.27	0.27	0.27	0.27	0.3	0.26	0.24	0.21	0.182	0.144	0.1					
41	0	0.05	0.1	0.142	0.18	0.21	0.237	0.256	0.27	0.27	0.27	0.27	0.3	0.26	0.24	0.21	0.181	0.142	0.098					
42	0	0.05	0.1	0.141	0.18	0.21	0.236	0.254	0.27	0.27	0.27	0.27	0.3	0.25	0.24	0.21	0.179	0.141	0.097					
43	0	0.05	0.1	0.139	0.18	0.21	0.234	0.252	0.26	0.27	0.27	0.27	0.3	0.25	0.23	0.21	0.177	0.139	0.096					
44	0	0.05	0.1	0.138	0.18	0.21	0.232	0.251	0.26	0.27	0.27	0.27	0.3	0.25	0.23	0.21	0.176	0.138	0.095					

47	0	0.047	0.09	0.13	0.171	0.2	0.2	0.25	0.26	0.26	0.264	0.3	0.245	0.227	0.202	0.171	0.134	0.0
48	0	0.047	0.09	0.13	0.169	0.2	0.2	0.24	0.26	0.26	0.262	0.3	0.244	0.225	0.2	0.169	0.133	0.0
49	0	0.046	0.09	0.13	0.168	0.2	0.2	0.24	0.25	0.26	0.261	0.3	0.242	0.223	0.199	0.168	0.131	0.0
50	0	0.046	0.09	0.13	0.166	0.2	0.2	0.24	0.25	0.26	0.259	0.3	0.24	0.222	0.197	0.166	0.13	0.0
51	0	0.045	0.09	0.13	0.165	0.2	0.2	0.24	0.25	0.26	0.257	0.3	0.239	0.22	0.195	0.165	0.129	0.0
52	0	0.045	0.09	0.13	0.163	0.19	0.2	0.24	0.25	0.26	0.256	0.2	0.237	0.218	0.194	0.163	0.128	0.0
53	0	0.044	0.09	0.13	0.162	0.19	0.2	0.24	0.25	0.25	0.254	0.2	0.235	0.217	0.192	0.162	0.127	0.0
54	0	0.044	0.09	0.13	0.161	0.19	0.2	0.23	0.25	0.25	0.252	0.2	0.234	0.215	0.191	0.161	0.125	0.0
55	0	0.043	0.09	0.12	0.159	0.19	0.2	0.23	0.24	0.25	0.251	0.2	0.232	0.214	0.189	0.159	0.124	0.0
56	0	0.043	0.08	0.12	0.158	0.19	0.2	0.23	0.24	0.25	0.249	0.2	0.23	0.212	0.188	0.158	0.123	0.0
57	0	0.043	0.08	0.12	0.157	0.19	0.2	0.23	0.24	0.25	0.247	0.2	0.229	0.21	0.186	0.157	0.122	0.0
58	0	0.042	0.08	0.12	0.155	0.18	0.2	0.23	0.24	0.25	0.246	0.2	0.227	0.209	0.185	0.155	0.121	0.0
59	0	0.042	0.08	0.12	0.154	0.18	0.2	0.23	0.24	0.24	0.244	0.2	0.226	0.207	0.183	0.154	0.12	0.0
60	0	0.041	0.08	0.12	0.153	0.18	0.2	0.22	0.24	0.24	0.242	0.2	0.224	0.206	0.182	0.153	0.119	0.0
61	0	0.041	0.08	0.12	0.152	0.18	0.2	0.22	0.23	0.24	0.241	0.2	0.222	0.204	0.181	0.152	0.118	0.0
62	0	0.041	0.08	0.12	0.15	0.18	0.2	0.22	0.23	0.24	0.239	0.2	0.221	0.203	0.179	0.15	0.117	0.0
63	0	0.04	0.08	0.12	0.149	0.18	0.2	0.22	0.23	0.24	0.237	0.2	0.219	0.201	0.178	0.149	0.116	0.0
64	0	0.04	0.08	0.12	0.148	0.18	0.2	0.22	0.23	0.24	0.236	0.2	0.218	0.2	0.176	0.148	0.115	0.0
65	0	0.04	0.08	0.11	0.147	0.18	0.2	0.22	0.23	0.23	0.234	0.2	0.216	0.198	0.175	0.147	0.114	0.0
66	0	0.039	0.08	0.11	0.146	0.17	0.2	0.21	0.23	0.23	0.233	0.2	0.215	0.197	0.174	0.146	0.113	0.0
67	0	0.039	0.08	0.11	0.144	0.17	0.2	0.21	0.22	0.23	0.231	0.2	0.213	0.195	0.172	0.144	0.112	0.0
68	0	0.039	0.08	0.11	0.143	0.17	0.2	0.21	0.22	0.23	0.229	0.2	0.211	0.194	0.171	0.143	0.111	0.0
69	0	0.038	0.08	0.11	0.142	0.17	0.2	0.21	0.22	0.23	0.228	0.2	0.21	0.192	0.17	0.142	0.111	0.0
70	0	0.038	0.08	0.11	0.141	0.17	0.2	0.21	0.22	0.23	0.226	0.2	0.208	0.191	0.168	0.141	0.11	0.0
71	0	0.038	0.07	0.11	0.14	0.17	0.2	0.21	0.22	0.22	0.225	0.2	0.207	0.19	0.167	0.14	0.109	0.0
72	0	0.037	0.07	0.11	0.139	0.17	0.2	0.21	0.22	0.22	0.223	0.2	0.205	0.188	0.166	0.139	0.108	0.0
73	0	0.037	0.07	0.11	0.138	0.16	0.2	0.2	0.22	0.22	0.221	0.2	0.204	0.187	0.165	0.138	0.107	0.0
74	0	0.037	0.07	0.11	0.137	0.16	0.2	0.21	0.22	0.22	0.22	0.2	0.202	0.186	0.163	0.137	0.106	0.0
75	0	0.037	0.07	0.11	0.136	0.16	0.2	0.21	0.22	0.218	0.2	0.201	0.184	0.162	0.136	0.105	0.0	
76	0	0.036	0.07	0.1	0.135	0.16	0.2	0.21	0.21	0.22	0.217	0.2	0.2	0.183	0.161	0.135	0.105	0.0
77	0	0.036	0.07	0.1	0.134	0.16	0.2	0.21	0.21	0.22	0.215	0.2	0.198	0.181	0.16	0.134	0.104	0.0
78	0	0.036	0.07	0.1	0.133	0.16	0.2	0.21	0.21	0.214	0.2	0.197	0.18	0.159	0.133	0.103	0.0	
79	0	0.035	0.07	0.1	0.132	0.16	0.2	0.21	0.21	0.212	0.2	0.195	0.179	0.157	0.132	0.102	0.0	
80	0	0.035	0.07	0.1	0.131	0.16	0.2	0.19	0.2	0.21	0.211	0.2	0.194	0.177	0.156	0.131	0.101	0.0
81	0	0.035	0.07	0.1	0.13	0.16	0.2	0.19	0.2	0.21	0.209	0.2	0.192	0.176	0.155	0.13	0.101	0.0
82	0	0.035	0.07	0.1	0.129	0.15	0.2	0.19	0.2	0.21	0.208	0.2	0.191	0.175	0.154	0.129	0.1	0.0
83	0	0.034	0.07	0.1	0.128	0.15	0.2	0.19	0.2	0.21	0.206	0.2	0.19	0.174	0.153	0.128	0.099	0.0
84	0	0.034	0.07	0.1	0.127	0.15	0.2	0.19	0.2	0.2	0.205	0.2	0.188	0.172	0.152	0.127	0.098	0.0
85	0	0.034	0.07	0.1	0.126	0.15	0.2	0.19	0.2	0.2	0.203	0.2	0.187	0.171	0.151	0.126	0.098	0.0
86	0	0.034	0.07	0.1	0.125	0.15	0.2	0.19	0.2	0.2	0.202	0.2	0.186	0.17	0.149	0.125	0.097	0.0
87	0	0.033	0.07	0.1	0.124	0.15	0.2	0.18	0.19	0.2	0.2	0.2	0.184	0.169	0.148	0.124	0.096	0.0
88	0	0.033	0.07	0.1	0.123	0.15	0.2	0.18	0.19	0.2	0.199	0.2	0.183	0.167	0.147	0.123	0.095	0.0
89	0	0.033	0.06	0.09	0.122	0.15	0.2	0.18	0.19	0.2	0.197	0.2	0.182	0.166	0.146	0.122	0.095	0.0
90	0	0.033	0.06	0.09	0.121	0.15	0.2	0.18	0.19	0.2	0.196	0.2	0.18	0.165	0.145	0.121	0.094	0.0
91	0	0.032	0.06	0.09	0.12	0.14	0.2	0.18	0.19	0.19	0.195	0.2	0.179	0.164	0.144	0.12	0.093	0.0

92	0	0.032	0.06	0.093	0.119	0.14	0.16	0.2	0.188	0.19	0.193	0.19	0.178	0.162	0.14	0.119	0.09	0.063
93	0	0.032	0.06	0.092	0.119	0.14	0.16	0.2	0.187	0.19	0.192	0.19	0.176	0.161	0.14	0.119	0.09	0.063
94	0	0.032	0.06	0.091	0.118	0.14	0.16	0.2	0.185	0.19	0.19	0.19	0.175	0.16	0.14	0.118	0.09	0.062
95	0	0.031	0.06	0.091	0.117	0.14	0.16	0.2	0.184	0.19	0.189	0.18	0.174	0.159	0.14	0.117	0.09	0.062
96	0	0.031	0.06	0.09	0.116	0.14	0.16	0.2	0.182	0.19	0.188	0.18	0.172	0.158	0.14	0.116	0.09	0.061
97	0	0.031	0.06	0.089	0.115	0.14	0.16	0.2	0.181	0.19	0.186	0.18	0.171	0.157	0.14	0.115	0.09	0.061
98	0	0.031	0.06	0.089	0.114	0.14	0.16	0.2	0.18	0.18	0.185	0.18	0.17	0.155	0.14	0.114	0.09	0.06
99	0	0.03	0.06	0.088	0.113	0.14	0.15	0.2	0.179	0.18	0.184	0.18	0.169	0.154	0.14	0.113	0.09	0.06
100	0	0.03	0.06	0.087	0.113	0.13	0.15	0.2	0.177	0.18	0.182	0.18	0.167	0.153	0.13	0.113	0.09	0.06
101	0	0.03	0.06	0.087	0.112	0.13	0.15	0.2	0.176	0.18	0.181	0.18	0.166	0.152	0.13	0.112	0.09	0.059
102	0	0.03	0.06	0.086	0.111	0.13	0.15	0.2	0.175	0.18	0.18	0.17	0.165	0.151	0.13	0.111	0.09	0.059
103	0	0.03	0.06	0.085	0.11	0.13	0.15	0.2	0.173	0.18	0.178	0.17	0.164	0.15	0.13	0.11	0.09	0.058
104	0	0.029	0.06	0.085	0.109	0.13	0.15	0.2	0.172	0.18	0.177	0.17	0.163	0.149	0.13	0.109	0.08	0.058
105	0	0.029	0.06	0.084	0.108	0.13	0.15	0.2	0.171	0.18	0.176	0.17	0.161	0.148	0.13	0.108	0.08	0.057
106	0	0.029	0.06	0.083	0.108	0.13	0.15	0.2	0.17	0.17	0.174	0.17	0.16	0.147	0.13	0.108	0.08	0.057
107	0	0.029	0.06	0.083	0.107	0.13	0.15	0.2	0.168	0.17	0.173	0.17	0.159	0.146	0.13	0.107	0.08	0.057
108	0	0.028	0.06	0.082	0.106	0.13	0.14	0.2	0.167	0.17	0.172	0.17	0.158	0.144	0.13	0.106	0.08	0.056
109	0	0.028	0.06	0.082	0.105	0.13	0.14	0.2	0.166	0.17	0.171	0.17	0.157	0.143	0.13	0.105	0.08	0.056
110	0	0.028	0.06	0.081	0.104	0.13	0.14	0.2	0.165	0.17	0.169	0.16	0.156	0.142	0.13	0.104	0.08	0.055
111	0	0.028	0.05	0.08	0.104	0.12	0.14	0.2	0.164	0.17	0.168	0.16	0.155	0.141	0.12	0.104	0.08	0.055
112	0	0.028	0.05	0.08	0.103	0.12	0.14	0.2	0.162	0.17	0.167	0.16	0.153	0.14	0.12	0.103	0.08	0.054
113	0	0.027	0.05	0.079	0.102	0.12	0.14	0.2	0.161	0.17	0.166	0.16	0.152	0.139	0.12	0.102	0.08	0.054
114	0	0.027	0.05	0.079	0.101	0.12	0.14	0.2	0.16	0.16	0.164	0.16	0.151	0.138	0.12	0.101	0.08	0.054
115	0	0.027	0.05	0.078	0.101	0.12	0.14	0.2	0.159	0.16	0.163	0.16	0.15	0.137	0.12	0.101	0.08	0.053
116	0	0.027	0.05	0.077	0.1	0.12	0.14	0.1	0.158	0.16	0.162	0.16	0.149	0.136	0.12	0.1	0.08	0.053
117	0	0.027	0.05	0.077	0.099	0.12	0.14	0.1	0.157	0.16	0.161	0.16	0.148	0.135	0.12	0.099	0.08	0.052
118	0	0.026	0.05	0.076	0.099	0.12	0.13	0.1	0.155	0.16	0.16	0.16	0.147	0.134	0.12	0.099	0.08	0.052
119	0	0.026	0.05	0.076	0.098	0.12	0.13	0.1	0.154	0.16	0.159	0.15	0.146	0.133	0.12	0.098	0.08	0.052
120	0	0.026	0.05	0.075	0.097	0.12	0.13	0.1	0.153	0.16	0.157	0.15	0.145	0.132	0.12	0.097	0.08	0.051
121	0	0.026	0.05	0.075	0.096	0.12	0.13	0.1	0.152	0.16	0.156	0.15	0.144	0.131	0.12	0.096	0.07	0.051
122	0	0.026	0.05	0.074	0.096	0.11	0.13	0.1	0.151	0.16	0.155	0.15	0.143	0.13	0.11	0.096	0.07	0.051
123	0	0.025	0.05	0.074	0.095	0.11	0.13	0.1	0.15	0.15	0.154	0.15	0.142	0.129	0.11	0.095	0.07	0.05
124	0	0.025	0.05	0.073	0.094	0.11	0.13	0.1	0.149	0.15	0.153	0.15	0.14	0.128	0.11	0.094	0.07	0.05
125	0	0.025	0.05	0.073	0.094	0.11	0.13	0.1	0.148	0.15	0.152	0.15	0.139	0.127	0.11	0.094	0.07	0.049
126	0	0.025	0.05	0.072	0.093	0.11	0.13	0.1	0.147	0.15	0.151	0.15	0.138	0.127	0.11	0.093	0.07	0.049
127	0	0.025	0.05	0.071	0.092	0.11	0.13	0.1	0.145	0.15	0.15	0.15	0.137	0.126	0.11	0.092	0.07	0.049
128	0	0.025	0.05	0.071	0.092	0.11	0.12	0.1	0.144	0.15	0.148	0.14	0.136	0.125	0.11	0.092	0.07	0.048
129	0	0.024	0.05	0.07	0.091	0.11	0.12	0.1	0.143	0.15	0.147	0.14	0.135	0.124	0.11	0.091	0.07	0.048
130	0	0.024	0.05	0.07	0.09	0.11	0.12	0.1	0.142	0.15	0.146	0.14	0.134	0.123	0.11	0.09	0.07	0.048
131	0	0.024	0.05	0.069	0.09	0.11	0.12	0.1	0.141	0.15	0.145	0.14	0.133	0.122	0.11	0.09	0.07	0.047
132	0	0.024	0.05	0.069	0.089	0.11	0.12	0.1	0.14	0.14	0.144	0.14	0.132	0.121	0.11	0.089	0.07	0.047
133	0	0.024	0.05	0.068	0.088	0.11	0.12	0.1	0.139	0.14	0.143	0.14	0.132	0.12	0.11	0.088	0.07	0.047
134	0	0.023	0.05	0.068	0.088	0.1	0.12	0.1	0.138	0.14	0.142	0.14	0.131	0.119	0.1	0.088	0.07	0.046
135	0	0.023	0.05	0.067	0.087	0.1	0.12	0.1	0.137	0.14	0.141	0.14	0.13	0.118	0.1	0.087	0.07	0.046
136	0	0.023	0.05	0.067	0.086	0.1	0.12	0.1	0.136	0.14	0.14	0.14	0.129	0.118	0.1	0.086	0.07	0.046
137	0	0.023	0.05	0.066	0.086	0.1	0.12	0.1	0.135	0.14	0.139	0.14	0.128	0.117	0.1	0.086	0.07	0.045

138	0	0.023	0.045	0.066	0.085	0.1	0.12	0.13	0.13	0.14	0.14	0.13	0.13	0.13	0.116	0.1	0.085	0.066	0.045
139	0	0.023	0.045	0.065	0.084	0.1	0.12	0.13	0.13	0.14	0.14	0.13	0.13	0.13	0.115	0.1	0.084	0.065	0.045
140	0	0.022	0.044	0.065	0.084	0.1	0.11	0.12	0.13	0.14	0.14	0.13	0.13	0.12	0.114	0.1	0.084	0.065	0.044
141	0	0.022	0.044	0.064	0.083	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.113	0.1	0.083	0.064	0.044
142	0	0.022	0.044	0.064	0.083	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.113	0.1	0.083	0.064	0.044
143	0	0.022	0.043	0.064	0.082	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.112	0.1	0.082	0.064	0.043
144	0	0.022	0.043	0.063	0.081	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.111	0.1	0.081	0.063	0.043
145	0	0.022	0.043	0.063	0.081	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.081	0.063	0.043
146	0	0.021	0.042	0.062	0.08	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.109	0.1	0.08	0.062	0.042
147	0	0.021	0.042	0.062	0.08	0.1	0.11	0.12	0.13	0.13	0.13	0.13	0.13	0.12	0.108	0.1	0.08	0.062	0.042
148	0	0.021	0.042	0.061	0.079	0.09	0.11	0.12	0.12	0.13	0.13	0.13	0.12	0.12	0.108	0.09	0.079	0.061	0.042
149	0	0.021	0.041	0.061	0.078	0.09	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.107	0.09	0.078	0.061	0.041	
150	0	0.021	0.041	0.06	0.078	0.09	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.106	0.09	0.078	0.06	0.041	
151	0	0.021	0.041	0.06	0.077	0.09	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.105	0.09	0.077	0.06	0.041	
152	0	0.021	0.041	0.059	0.077	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.105	0.09	0.077	0.059	0.041
153	0	0.02	0.04	0.059	0.076	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.104	0.09	0.076	0.059	0.04
154	0	0.02	0.04	0.059	0.076	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.103	0.09	0.076	0.059	0.04
155	0	0.02	0.04	0.058	0.075	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.102	0.09	0.075	0.058	0.04
156	0	0.02	0.039	0.058	0.075	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.102	0.09	0.075	0.058	0.039
157	0	0.02	0.039	0.057	0.074	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.101	0.09	0.074	0.057	0.039
158	0	0.02	0.039	0.057	0.073	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.073	0.057	0.039
159	0	0.02	0.039	0.056	0.073	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.12	0.11	0.099	0.09	0.073	0.056	0.039
160	0	0.019	0.038	0.056	0.072	0.09	0.1	0.11	0.11	0.12	0.12	0.12	0.11	0.11	0.099	0.09	0.072	0.056	0.038
161	0	0.019	0.038	0.056	0.072	0.09	0.1	0.11	0.11	0.12	0.12	0.12	0.11	0.11	0.098	0.09	0.072	0.056	0.038
162	0	0.019	0.038	0.055	0.071	0.09	0.1	0.11	0.11	0.12	0.12	0.12	0.11	0.11	0.097	0.09	0.071	0.055	0.038
163	0	0.019	0.037	0.055	0.071	0.08	0.1	0.11	0.11	0.11	0.11	0.11	0.11	0.11	0.096	0.08	0.071	0.055	0.037
164	0	0.019	0.037	0.054	0.07	0.08	0.1	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.096	0.08	0.07	0.054	0.037
165	0	0.019	0.037	0.054	0.07	0.08	0.1	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.095	0.08	0.07	0.054	0.037
166	0	0.019	0.037	0.054	0.069	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.094	0.08	0.069	0.054	0.037
167	0	0.018	0.036	0.053	0.069	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.094	0.08	0.069	0.053	0.036
168	0	0.018	0.036	0.053	0.068	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.093	0.08	0.068	0.053	0.036
169	0	0.018	0.036	0.052	0.068	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.092	0.08	0.068	0.052	0.036
170	0	0.018	0.036	0.052	0.067	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.092	0.08	0.067	0.052	0.036
171	0	0.018	0.035	0.052	0.067	0.08	0.09	0.1	0.11	0.11	0.11	0.11	0.11	0.1	0.091	0.08	0.067	0.052	0.035
172	0	0.018	0.035	0.051	0.066	0.08	0.09	0.1	0.1	0.11	0.11	0.11	0.1	0.1	0.09	0.08	0.066	0.051	0.035
173	0	0.018	0.035	0.051	0.066	0.08	0.09	0.1	0.1	0.11	0.11	0.11	0.1	0.1	0.09	0.08	0.066	0.051	0.035
174	0	0.017	0.035	0.051	0.065	0.08	0.09	0.1	0.1	0.11	0.11	0.11	0.1	0.1	0.089	0.08	0.065	0.051	0.035
175	0	0.017	0.034	0.05	0.065	0.08	0.09	0.1	0.1	0.11	0.11	0.11	0.1	0.1	0.088	0.08	0.065	0.05	0.034
176	0	0.017	0.034	0.05	0.064	0.08	0.09	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.088	0.08	0.064	0.05	0.034
177	0	0.017	0.034	0.049	0.064	0.08	0.09	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.087	0.08	0.064	0.049	0.034
178	0	0.017	0.034	0.049	0.063	0.08	0.09	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.086	0.08	0.063	0.049	0.034
179	0	0.017	0.033	0.049	0.063	0.08	0.09	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.086	0.08	0.063	0.049	0.033
180	0	0.017	0.033	0.048	0.062	0.07	0.09	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.085	0.07	0.062	0.048	0.033
181	0	0.017	0.033	0.048	0.062	0.07	0.08	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.085	0.07	0.062	0.048	0.033
182	0	0.016	0.033	0.048	0.062	0.07	0.08	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.084	0.07	0.062	0.048	0.033
183	0	0.016	0.032	0.047	0.061	0.07	0.08	0.09	0.1	0.1	0.1	0.1	0.1	0.09	0.083	0.07	0.061	0.047	0.032

184	0	0.016	0.03	0.05	0.06	0.073	0.083	0.09	0.096	0.1	0.1	0.1	0.09	0.1	0.073	0.0607	0.05	0.032
185	0	0.016	0.03	0.05	0.06	0.072	0.082	0.09	0.095	0.1	0.1	0.1	0.09	0.1	0.072	0.0602	0.05	0.032
186	0	0.016	0.03	0.05	0.06	0.072	0.081	0.09	0.094	0.1	0.1	0.09	0.089	0.1	0.072	0.0598	0.05	0.032
187	0	0.016	0.03	0.05	0.06	0.071	0.081	0.09	0.094	0.1	0.1	0.09	0.088	0.1	0.071	0.0593	0.05	0.031
188	0	0.016	0.03	0.05	0.06	0.071	0.08	0.09	0.093	0.1	0.1	0.09	0.088	0.1	0.071	0.0589	0.05	0.031
189	0	0.016	0.03	0.05	0.06	0.07	0.08	0.09	0.092	0.09	0.09	0.09	0.087	0.1	0.07	0.0585	0.05	0.031
190	0	0.016	0.03	0.04	0.06	0.07	0.079	0.09	0.092	0.09	0.09	0.09	0.087	0.1	0.07	0.0581	0.04	0.031
191	0	0.015	0.03	0.04	0.06	0.069	0.079	0.09	0.091	0.09	0.09	0.09	0.086	0.1	0.069	0.0576	0.04	0.03
192	0	0.015	0.03	0.04	0.06	0.069	0.078	0.09	0.09	0.09	0.09	0.09	0.085	0.1	0.069	0.0572	0.04	0.03
193	0	0.015	0.03	0.04	0.06	0.068	0.077	0.08	0.09	0.09	0.09	0.09	0.085	0.1	0.068	0.0568	0.04	0.03
194	0	0.015	0.03	0.04	0.06	0.068	0.077	0.08	0.089	0.09	0.09	0.09	0.084	0.1	0.068	0.0564	0.04	0.03
195	0	0.015	0.03	0.04	0.06	0.067	0.076	0.08	0.088	0.09	0.09	0.09	0.083	0.1	0.067	0.056	0.04	0.03
196	0	0.015	0.03	0.04	0.06	0.067	0.076	0.08	0.088	0.09	0.09	0.09	0.083	0.1	0.067	0.0556	0.04	0.029
197	0	0.015	0.03	0.04	0.06	0.066	0.075	0.08	0.087	0.09	0.09	0.09	0.082	0.1	0.066	0.0551	0.04	0.029
198	0	0.015	0.03	0.04	0.05	0.066	0.075	0.08	0.086	0.09	0.09	0.09	0.082	0.1	0.066	0.0547	0.04	0.029
199	0	0.015	0.03	0.04	0.05	0.065	0.074	0.08	0.086	0.09	0.09	0.09	0.081	0.1	0.065	0.0543	0.04	0.029
200	0	0.014	0.03	0.04	0.05	0.065	0.074	0.08	0.085	0.09	0.09	0.09	0.08	0.1	0.065	0.0539	0.04	0.029
201	0	0.014	0.03	0.04	0.05	0.064	0.073	0.08	0.085	0.09	0.09	0.08	0.08	0.1	0.064	0.0536	0.04	0.028
202	0	0.014	0.03	0.04	0.05	0.064	0.072	0.08	0.084	0.09	0.09	0.08	0.079	0.1	0.064	0.0532	0.04	0.028
203	0	0.014	0.03	0.04	0.05	0.063	0.072	0.08	0.083	0.09	0.09	0.08	0.079	0.1	0.063	0.0528	0.04	0.028
204	0	0.014	0.03	0.04	0.05	0.063	0.071	0.08	0.083	0.08	0.08	0.08	0.078	0.1	0.063	0.0524	0.04	0.028
205	0	0.014	0.03	0.04	0.05	0.062	0.071	0.08	0.082	0.08	0.08	0.08	0.078	0.1	0.062	0.052	0.04	0.027
206	0	0.014	0.03	0.04	0.05	0.062	0.07	0.08	0.081	0.08	0.08	0.08	0.077	0.1	0.062	0.0516	0.04	0.027
207	0	0.014	0.03	0.04	0.05	0.061	0.07	0.08	0.081	0.08	0.08	0.08	0.076	0.1	0.061	0.0512	0.04	0.027
208	0	0.014	0.03	0.04	0.05	0.061	0.069	0.08	0.08	0.08	0.08	0.08	0.076	0.1	0.061	0.0509	0.04	0.027
209	0	0.014	0.03	0.04	0.05	0.06	0.069	0.08	0.08	0.08	0.08	0.08	0.075	0.1	0.06	0.0505	0.04	0.027
210	0	0.013	0.03	0.04	0.05	0.06	0.068	0.07	0.079	0.08	0.08	0.08	0.075	0.1	0.06	0.0501	0.04	0.027
211	0	0.013	0.03	0.04	0.05	0.06	0.068	0.07	0.079	0.08	0.08	0.08	0.074	0.1	0.06	0.0498	0.04	0.026
212	0	0.013	0.03	0.04	0.05	0.059	0.067	0.07	0.078	0.08	0.08	0.08	0.074	0.1	0.059	0.0494	0.04	0.026
213	0	0.013	0.03	0.04	0.05	0.059	0.067	0.07	0.077	0.08	0.08	0.08	0.073	0.1	0.059	0.049	0.04	0.026
214	0	0.013	0.03	0.04	0.05	0.058	0.066	0.07	0.077	0.08	0.08	0.08	0.073	0.1	0.058	0.0487	0.04	0.026
215	0	0.013	0.03	0.04	0.05	0.058	0.066	0.07	0.076	0.08	0.08	0.08	0.072	0.1	0.058	0.0483	0.04	0.026
216	0	0.013	0.03	0.04	0.05	0.057	0.065	0.07	0.076	0.08	0.08	0.08	0.072	0.1	0.057	0.048	0.04	0.025
217	0	0.013	0.03	0.04	0.05	0.057	0.065	0.07	0.075	0.08	0.08	0.08	0.071	0.1	0.057	0.0476	0.04	0.025
218	0	0.013	0.02	0.04	0.05	0.057	0.064	0.07	0.075	0.08	0.08	0.07	0.07	0.1	0.057	0.0473	0.04	0.025
219	0	0.013	0.02	0.04	0.05	0.056	0.064	0.07	0.074	0.08	0.08	0.07	0.07	0.1	0.056	0.0469	0.04	0.025
220	0	0.012	0.02	0.04	0.05	0.056	0.063	0.07	0.074	0.08	0.08	0.07	0.069	0.1	0.056	0.0466	0.04	0.025
221	0	0.012	0.02	0.04	0.05	0.055	0.063	0.07	0.073	0.08	0.08	0.07	0.069	0.1	0.055	0.0462	0.04	0.024
222	0	0.012	0.02	0.04	0.05	0.055	0.063	0.07	0.072	0.07	0.07	0.07	0.068	0.1	0.055	0.0459	0.04	0.024
223	0	0.012	0.02	0.04	0.05	0.055	0.062	0.07	0.072	0.07	0.07	0.07	0.068	0.1	0.055	0.0456	0.04	0.024
224	0	0.012	0.02	0.04	0.05	0.054	0.062	0.07	0.071	0.07	0.07	0.07	0.067	0.1	0.054	0.0452	0.04	0.024
225	0	0.012	0.02	0.03	0.04	0.054	0.061	0.07	0.071	0.07	0.07	0.07	0.067	0.1	0.054	0.0449	0.03	0.024
226	0	0.012	0.02	0.03	0.04	0.053	0.061	0.07	0.07	0.07	0.07	0.07	0.066	0.1	0.053	0.0446	0.03	0.024
227	0	0.012	0.02	0.03	0.04	0.053	0.06	0.07	0.07	0.07	0.07	0.07	0.066	0.1	0.053	0.0443	0.03	0.023
228	0	0.012	0.02	0.03	0.04	0.053	0.06	0.07	0.069	0.07	0.07	0.07	0.065	0.1	0.053	0.0439	0.03	0.023



274	0	0.008	0.017	0.024	0.031	0.04	0.04	0.05	0.049	0.051	0.051	0.05	0.047	0.043	0.04	0.031	0.02	0.0
275	0	0.008	0.016	0.024	0.031	0.04	0.04	0.05	0.049	0.05	0.05	0.05	0.046	0.042	0.04	0.031	0.02	0.0
276	0	0.008	0.016	0.024	0.031	0.04	0.04	0.05	0.049	0.05	0.05	0.05	0.046	0.042	0.04	0.031	0.02	0.0
277	0	0.008	0.016	0.024	0.031	0.04	0.04	0.05	0.048	0.05	0.05	0.05	0.046	0.042	0.04	0.031	0.02	0.0
278	0	0.008	0.016	0.024	0.03	0.04	0.04	0.05	0.048	0.049	0.049	0.05	0.045	0.041	0.04	0.03	0.02	0.0
279	0	0.008	0.016	0.023	0.03	0.04	0.04	0.05	0.048	0.049	0.049	0.05	0.045	0.041	0.04	0.03	0.02	0.0
280	0	0.008	0.016	0.023	0.03	0.04	0.04	0.04	0.047	0.049	0.049	0.05	0.045	0.041	0.04	0.03	0.02	0.0

### B.3.3 Spreadsheet for the mass of oil leached at 99°C

Segments	18			
$\Delta x$	2.78E-05 m		Total mass	20 kg
$C_0$	1.68E+00 mol/m <sup>3</sup>		Density	960 kg/m <sup>3</sup>
$D$	8.74 E-12 m <sup>2</sup> /sec		Total Volume	2.08E-02 m <sup>3</sup>
$\Delta t$	30 sec		Thickness	0.0005 m
SSE:	5.11 E-01		Total area	41.66667 m <sup>2</sup>

#### B.3.3.1 Concentration (mol/m<sup>3</sup>) for the mass oil in the leaf at time $\Delta t$

$\Delta t$		Right edge	Leaf	Left edge

0	0	0	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	0		
30	1	0	1.1	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.1	0	
60	2	0	0.9	1.5	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.5	0.9	0	
90	3	0	0.8	1.4	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.4	0.8	0	
120	4	0	0.7	1.3	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.3	0.7	0	
150	5	0	0.7	1.2	1.5	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.2	0.7	0	
180	6	0	0.6	1.1	1.4	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.4	1.1	0.6	0	
210	7	0	0.6	1	1.4	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.4	1	0.6	0
240	8	0	0.5	1	1.3	1.5	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.3	1	0.5	0
270	9	0	0.5	1	1.3	1.5	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.3	1	0.5	0
300	10	0	0.5	0.9	1.2	1.5	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.2	0.9	0.5	0
330	11	0	0.5	0.9	1.2	1.4	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.4	1.2	0.9	0.5	0
360	12	0	0.4	0.9	1.2	1.4	1.5	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.5	1.4	1.2	0.9	0.4	0
390	13	0	0.4	0.8	1.1	1.4	1.5	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.4	1.1	0.8	0.4	0	0	0	
420	14	0	0.4	0.8	1.1	1.3	1.5	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0	0	0	0	
450	15	0	0.4	0.8	1.1	1.3	1.5	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0	0	0	0	
480	16	0	0.4	0.8	1.1	1.3	1.5	1.6	1.6	1.7	1.7	1.7	1.7	1.7	1.6	1.6	1.5	1.3	1.1	0.8	0.4	0	0	0	0	
510	17	0	0.4	0.7	1	1.3	1.4	1.5	1.6	1.6	1.7	1.7	1.7	1.6	1.6	1.5	1.4	1.3	1	0.7	0.4	0	0	0	0	
540	18	0	0.4	0.7	1	1.2	1.4	1.5	1.6	1.6	1.7	1.7	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0	0	0	0	0	
570	19	0	0.4	0.7	1	1.2	1.4	1.5	1.6	1.6	1.7	1.7	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0	0	0	0	0	
600	20	0	0.4	0.7	1	1.2	1.4	1.5	1.6	1.6	1.6	1.6	1.6	1.6	1.5	1.4	1.2	1	0.7	0.4	0	0	0	0	0	
630	21	0	0.3	0.7	1	1.2	1.4	1.5	1.6	1.6	1.6	1.6	1.6	1.6	1.5	1.4	1.2	1	0.7	0.3	0	0	0	0	0	
660	22	0	0.3	0.7	0.9	1.2	1.3	1.5	1.6	1.6	1.6	1.6	1.6	1.6	1.5	1.3	1.2	0.9	0.7	0.3	0	0	0	0	0	
690	23	0	0.3	0.6	0.9	1.1	1.3	1.5	1.5	1.6	1.6	1.6	1.6	1.5	1.5	1.3	1.1	0.9	0.6	0.3	0	0	0	0	0	
720	24	0	0.3	0.6	0.9	1.1	1.3	1.4	1.5	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0	0	0	0	0	
750	25	0	0.3	0.6	0.9	1.1	1.3	1.4	1.5	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0	0	0	0	0	
780	26	0	0.3	0.6	0.9	1.1	1.3	1.4	1.5	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0	0	0	0	0	
810	27	0	0.3	0.6	0.9	1.1	1.3	1.4	1.5	1.6	1.6	1.6	1.6	1.5	1.4	1.3	1.1	0.9	0.6	0.3	0	0	0	0	0	
840	28	0	0.3	0.6	0.8	1.1	1.2	1.4	1.5	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1.1	0.8	0.6	0.3	0	0	0	0	0	
870	29	0	0.3	0.6	0.8	1.1	1.2	1.4	1.5	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1.1	0.8	0.6	0.3	0	0	0	0	0	
900	30	0	0.3	0.6	0.8	1	1.2	1.4	1.5	1.5	1.6	1.6	1.5	1.5	1.4	1.2	1	0.8	0.6	0.3	0	0	0	0	0	
930	31	0	0.3	0.6	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.6	0.3	0	0	0	0	0	
960	32	0	0.3	0.6	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.6	0.3	0	0	0	0	0	
990	33	0	0.3	0.5	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0	0	0	0	0	
1020	34	0	0.3	0.5	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0	0	0	0	0	
1050	35	0	0.3	0.5	0.8	1	1.2	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.2	1	0.8	0.5	0.3	0	0	0	0	0	
1080	36	0	0.3	0.5	0.8	1	1.1	1.3	1.4	1.5	1.5	1.5	1.5	1.4	1.3	1.1	1	0.8	0.5	0.3	0	0	0	0	0	
1110	37	0	0.3	0.5	0.7	1	1.1	1.3	1.4	1.4	1.5	1.5	1.4	1.4	1.3	1.1	1	0.7	0.5	0.3	0	0	0	0	0	
1140	38	0	0.3	0.5	0.7	0.9	1.1	1.3	1.4	1.4	1.5	1.5	1.4	1.4	1.3	1.1	0.9	0.7	0.5	0.3	0	0	0	0	0	
1170	39	0	0.3	0.5	0.7	0.9	1.1	1.2	1.4	1.4	1.5	1.5	1.4	1.4	1.2	1.1	0.9	0.7	0.5	0.3	0	0	0	0	0	
1200	40	0	0.3	0.5	0.7	0.9	1.1	1.2	1.3	1.4	1.4	1.4	1.4	1.3	1.2	1.1	0.9	0.7	0.5	0.3	0	0	0	0	0	
1230	41	0	0.2	0.5	0.7	0.9	1.1	1.2	1.3	1.4	1.4	1.4	1.4	1.3	1.2	1.1	0.9	0.7	0.5	0.2	0	0	0	0	0	
1260	42	0	0.2	0.5	0.7	0.9	1.1	1.2	1.3	1.4	1.4	1.4	1.4	1.3	1.2	1.1	0.9	0.7	0.5	0.2	0	0	0	0	0	
1290	43	0	0.2	0.5	0.7	0.9	1.1	1.2	1.3	1.4	1.4	1.4	1.4	1.3	1.2	1.1	0.9	0.7	0.5	0.2	0	0	0	0	0	
1320	44	0	0.2	0.5	0.7	0.9	1.1	1.2	1.3	1.4	1.4	1.4	1.4	1.3	1.2	1.1	0.9	0.7	0.5	0.2	0	0	0	0	0	
1350	45	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.2	1	0.9	0.7	0.5	0.2	0	0	0	0	0	
1380	46	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.3	1.3	1.2	1	0.9	0.7	0.5	0.2	0	0	0	0	0	

1410	47	0	0.2	0.5	0.7	0.9	1	1.2	1.3	1.3	1.4	1.4	1.33	1.3	1.2	1	0.9	0.7	0.46	0.2	0	
1440	48	0	0.2	0.5	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.31	1.2	1.1	1	0.8	0.7	0.45	0.2	0	
1470	49	0	0.2	0.4	0.7	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.29	1.2	1.1	1	0.8	0.7	0.45	0.2	0
1500	50	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.29	1.2	1.1	1	0.8	0.6	0.44	0.2	0
1530	51	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.28	1.2	1.1	1	0.8	0.6	0.44	0.2	0
1560	52	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.27	1.2	1.1	1	0.8	0.6	0.43	0.2	0
1590	53	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.3	1.3	1.3	1.3	1.26	1.2	1.1	1	0.8	0.6	0.43	0.2	0
1620	54	0	0.2	0.4	0.6	0.8	1	1.1	1.2	1.2	1.3	1.3	1.3	1.25	1.2	1.1	1	0.8	0.6	0.42	0.2	0
1650	55	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.23	1.2	1.1	0.9	0.8	0.6	0.42	0.2	0	
1680	56	0	0.2	0.4	0.6	0.8	0.9	1.1	1.2	1.2	1.3	1.3	1.22	1.2	1.1	0.9	0.8	0.6	0.42	0.2	0	
1710	57	0	0.2	0.4	0.6	0.8	0.9	1.1	1.1	1.2	1.2	1.2	1.21	1.1	1.1	0.9	0.8	0.6	0.41	0.2	0	
1740	58	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.2	1.1	1	0.9	0.8	0.6	0.41	0.2	0	
1770	59	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.19	1.1	1	0.9	0.8	0.6	0.4	0.2	0	
1800	60	0	0.2	0.4	0.6	0.8	0.9	1	1.1	1.2	1.2	1.2	1.18	1.1	1	0.9	0.8	0.6	0.4	0.2	0	
1830	61	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.2	1.2	1.2	1.17	1.1	1	0.9	0.7	0.6	0.4	0.2	0	
1860	62	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.2	1.2	1.2	1.16	1.1	1	0.9	0.7	0.6	0.39	0.2	0	
1890	63	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.1	1.2	1.2	1.15	1.1	1	0.9	0.7	0.6	0.39	0.2	0	
1920	64	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.1	1.2	1.2	1.14	1.1	1	0.9	0.7	0.6	0.38	0.2	0	
1950	65	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.1	1.2	1.2	1.13	1.1	1	0.9	0.7	0.6	0.38	0.2	0	
1980	66	0	0.2	0.4	0.6	0.7	0.9	1	1.1	1.1	1.1	1.1	1.12	1.1	1	0.9	0.7	0.6	0.38	0.2	0	
2010	67	0	0.2	0.4	0.5	0.7	0.8	1	1	1.1	1.1	1.1	1.11	1	1	0.8	0.7	0.5	0.37	0.2	0	
2040	68	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1.1	1.1	1.1	1.1	1	0.9	0.8	0.7	0.5	0.37	0.2	0	
2070	69	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1.1	1.1	1.1	1.09	1	0.9	0.8	0.7	0.5	0.37	0.2	0	
2100	70	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1.1	1.1	1.1	1.08	1	0.9	0.8	0.7	0.5	0.36	0.2	0	
2130	71	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1.1	1.1	1.1	1.07	1	0.9	0.8	0.7	0.5	0.36	0.2	0	
2160	72	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1.1	1.1	1.1	1.06	1	0.9	0.8	0.7	0.5	0.36	0.2	0	
2190	73	0	0.2	0.4	0.5	0.7	0.8	0.9	1	1	1.1	1.1	1.05	1	0.9	0.8	0.7	0.5	0.35	0.2	0	
2220	74	0	0.2	0.3	0.5	0.7	0.8	0.9	1	1	1.1	1.1	1.04	1	0.9	0.8	0.7	0.5	0.35	0.2	0	
2250	75	0	0.2	0.3	0.5	0.7	0.8	0.9	1	1	1.1	1.1	1.03	1	0.9	0.8	0.7	0.5	0.35	0.2	0	
2280	76	0	0.2	0.3	0.5	0.6	0.8	0.9	1	1	1	1	1.02	1	0.9	0.8	0.6	0.5	0.34	0.2	0	
2310	77	0	0.2	0.3	0.5	0.6	0.8	0.9	1	1	1	1	1.01	1	0.9	0.8	0.6	0.5	0.34	0.2	0	
2340	78	0	0.2	0.3	0.5	0.6	0.8	0.9	0.9	1	1	1	1	1	0.9	0.9	0.8	0.6	0.5	0.34	0.2	0
2370	79	0	0.2	0.3	0.5	0.6	0.8	0.9	0.9	1	1	1	1	0.99	0.9	0.9	0.8	0.6	0.5	0.33	0.2	0
2400	80	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	1	1	1	1	0.98	0.9	0.8	0.7	0.6	0.5	0.33	0.2	0
2430	81	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	1	1	1	1	0.97	0.9	0.8	0.7	0.6	0.5	0.33	0.2	0
2460	82	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	1	1	1	1	0.96	0.9	0.8	0.7	0.6	0.5	0.32	0.2	0
2490	83	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	1	1	1	1	0.95	0.9	0.8	0.7	0.6	0.5	0.32	0.2	0
2520	84	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	0.9	1	1	1	0.95	0.9	0.8	0.7	0.6	0.5	0.32	0.2	0
2550	85	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	0.9	1	1	1	0.94	0.9	0.8	0.7	0.6	0.5	0.31	0.2	0
2580	86	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	0.9	1	1	1	0.93	0.9	0.8	0.7	0.6	0.5	0.31	0.2	0
2610	87	0	0.2	0.3	0.5	0.6	0.7	0.8	0.9	0.9	0.9	0.9	0.92	0.9	0.8	0.7	0.6	0.5	0.31	0.2	0	
2640	88	0	0.2	0.3	0.4	0.6	0.7	0.8	0.9	0.9	0.9	0.9	0.91	0.9	0.8	0.7	0.6	0.4	0.31	0.2	0	
2670	89	0	0.2	0.3	0.4	0.6	0.7	0.8	0.9	0.9	0.9	0.9	0.9	0.8	0.7	0.6	0.4	0.3	0.2	0	0	

700	90	0	0.2	0.3	0.4	0.6	0.7	0.8	0.85	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.57	0.4	0.3	0.2	0
730	91	0	0.2	0.3	0.4	0.6	0.7	0.8	0.84	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.56	0.4	0.3	0.2	0
760	92	0	0.1	0.3	0.4	0.6	0.7	0.8	0.83	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.56	0.4	0.3	0.1	0
790	93	0	0.1	0.3	0.4	0.6	0.7	0.8	0.82	0.9	0.9	0.9	0.9	0.8	0.8	0.7	0.55	0.4	0.3	0.1	0
820	94	0	0.1	0.3	0.4	0.5	0.7	0.7	0.81	0.9	0.9	0.9	0.9	0.8	0.7	0.7	0.55	0.4	0.3	0.1	0
850	95	0	0.1	0.3	0.4	0.5	0.6	0.7	0.81	0.9	0.9	0.9	0.9	0.8	0.7	0.6	0.54	0.4	0.3	0.1	0
880	96	0	0.1	0.3	0.4	0.5	0.6	0.7	0.8	0.8	0.9	0.9	0.8	0.8	0.7	0.6	0.54	0.4	0.3	0.1	0
910	97	0	0.1	0.3	0.4	0.5	0.6	0.7	0.79	0.8	0.9	0.9	0.8	0.8	0.7	0.6	0.53	0.4	0.3	0.1	0
940	98	0	0.1	0.3	0.4	0.5	0.6	0.7	0.78	0.8	0.9	0.9	0.8	0.8	0.7	0.6	0.53	0.4	0.3	0.1	0
970	99	0	0.1	0.3	0.4	0.5	0.6	0.7	0.78	0.8	0.8	0.8	0.8	0.8	0.7	0.6	0.52	0.4	0.3	0.1	0
1000	100	0	0.1	0.3	0.4	0.5	0.6	0.7	0.77	0.8	0.8	0.8	0.8	0.8	0.7	0.6	0.52	0.4	0.3	0.1	0
1030	101	0	0.1	0.3	0.4	0.5	0.6	0.7	0.76	0.8	0.8	0.8	0.8	0.8	0.7	0.6	0.51	0.4	0.3	0.1	0
1060	102	0	0.1	0.3	0.4	0.5	0.6	0.7	0.76	0.8	0.8	0.8	0.8	0.8	0.7	0.6	0.51	0.4	0.3	0.1	0
1090	103	0	0.1	0.3	0.4	0.5	0.6	0.7	0.75	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.5	0.4	0.3	0.1	0
1120	104	0	0.1	0.3	0.4	0.5	0.6	0.7	0.74	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.5	0.4	0.3	0.1	0
1150	105	0	0.1	0.3	0.4	0.5	0.6	0.7	0.73	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.49	0.4	0.3	0.1	0
1180	106	0	0.1	0.3	0.4	0.5	0.6	0.7	0.73	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.49	0.4	0.3	0.1	0
1210	107	0	0.1	0.3	0.4	0.5	0.6	0.7	0.72	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.48	0.4	0.3	0.1	0
1240	108	0	0.1	0.3	0.4	0.5	0.6	0.7	0.71	0.8	0.8	0.8	0.8	0.7	0.7	0.6	0.48	0.4	0.3	0.1	0
1270	109	0	0.1	0.3	0.4	0.5	0.6	0.6	0.71	0.7	0.8	0.8	0.7	0.7	0.6	0.6	0.48	0.4	0.3	0.1	0
1300	110	0	0.1	0.2	0.4	0.5	0.6	0.6	0.7	0.7	0.8	0.8	0.7	0.7	0.6	0.6	0.47	0.4	0.2	0.1	0
1330	111	0	0.1	0.2	0.4	0.5	0.6	0.6	0.7	0.7	0.8	0.8	0.7	0.7	0.6	0.6	0.47	0.4	0.2	0.1	0
1360	112	0	0.1	0.2	0.4	0.5	0.6	0.6	0.69	0.7	0.7	0.7	0.7	0.7	0.6	0.6	0.46	0.4	0.2	0.1	0
1390	113	0	0.1	0.2	0.4	0.5	0.6	0.68	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.46	0.4	0.2	0.1	0	
1420	114	0	0.1	0.2	0.4	0.5	0.6	0.68	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.45	0.4	0.2	0.1	0	
1450	115	0	0.1	0.2	0.3	0.4	0.5	0.6	0.67	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.45	0.3	0.2	0.1	0
1480	116	0	0.1	0.2	0.3	0.4	0.5	0.6	0.66	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.45	0.3	0.2	0.1	0
1510	117	0	0.1	0.2	0.3	0.4	0.5	0.6	0.66	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.44	0.3	0.2	0.1	0
1540	118	0	0.1	0.2	0.3	0.4	0.5	0.6	0.65	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.44	0.3	0.2	0.1	0
1570	119	0	0.1	0.2	0.3	0.4	0.5	0.6	0.65	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.43	0.3	0.2	0.1	0
1600	120	0	0.1	0.2	0.3	0.4	0.5	0.6	0.64	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.43	0.3	0.2	0.1	0
1630	121	0	0.1	0.2	0.3	0.4	0.5	0.6	0.63	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.42	0.3	0.2	0.1	0
1660	122	0	0.1	0.2	0.3	0.4	0.5	0.6	0.63	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.42	0.3	0.2	0.1	0
1690	123	0	0.1	0.2	0.3	0.4	0.5	0.6	0.62	0.7	0.7	0.7	0.7	0.6	0.6	0.5	0.42	0.3	0.2	0.1	0
1720	124	0	0.1	0.2	0.3	0.4	0.5	0.6	0.62	0.7	0.7	0.7	0.7	0.6	0.6	0.5	0.41	0.3	0.2	0.1	0
1750	125	0	0.1	0.2	0.3	0.4	0.5	0.6	0.61	0.6	0.7	0.7	0.6	0.6	0.6	0.5	0.41	0.3	0.2	0.1	0
1780	126	0	0.1	0.2	0.3	0.4	0.5	0.6	0.6	0.6	0.7	0.7	0.6	0.6	0.6	0.5	0.41	0.3	0.2	0.1	0
1810	127	0	0.1	0.2	0.3	0.4	0.5	0.6	0.6	0.6	0.7	0.7	0.6	0.6	0.5	0.5	0.4	0.3	0.2	0.1	0
1840	128	0	0.1	0.2	0.3	0.4	0.5	0.5	0.59	0.6	0.6	0.6	0.6	0.6	0.6	0.5	0.4	0.3	0.2	0.1	0
1870	129	0	0.1	0.2	0.3	0.4	0.5	0.5	0.59	0.6	0.6	0.6	0.6	0.6	0.6	0.5	0.39	0.3	0.2	0.1	0
1900	130	0	0.1	0.2	0.3	0.4	0.5	0.5	0.58	0.6	0.6	0.6	0.6	0.6	0.5	0.5	0.39	0.3	0.2	0.1	0
1930	131	0	0.1	0.2	0.3	0.4	0.5	0.5	0.58	0.6	0.6	0.6	0.6	0.6	0.5	0.5	0.39	0.3	0.2	0.1	0
1960	132	0	0.1	0.2	0.3	0.4	0.5	0.5	0.57	0.6	0.6	0.6	0.6	0.6	0.5	0.5	0.38	0.3	0.2	0.1	0
1990	133	0	0.1	0.2	0.3	0.4	0.5	0.5	0.57	0.6	0.6	0.6	0.6	0.6	0.5	0.5	0.38	0.3	0.2	0.1	0







### B.3.3.2 Mass of oil leached

At		Right edge	Leaf	Left edge



0	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.
0	0.198	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.1980
0	0.165	0.27	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.1653
0	0.143	0.24	0.29	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.288	0.2	0.1431
0	0.128	0.22	0.28	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.277	0.2	0.1284
0	0.117	0.21	0.27	0.29	0.299	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.29	0.266	0.2	0.1174		
0	0.109	0.2	0.26	0.28	0.296	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.28	0.255	0.2	0.1088		
0	0.102	0.19	0.25	0.28	0.294	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.299	0.29	0.28	0.246	0.2	0.1019			
0	0.096	0.18	0.24	0.27	0.29	0.3	0.299	0.3	0.3	0.3	0.3	0.3	0.299	0.297	0.29	0.27	0.237	0.2	0.0961					
0	0.091	0.17	0.23	0.27	0.287	0.3	0.299	0.3	0.3	0.3	0.3	0.3	0.299	0.296	0.29	0.27	0.229	0.2	0.091					
0	0.087	0.16	0.22	0.26	0.283	0.29	0.298	0.3	0.3	0.3	0.3	0.3	0.298	0.294	0.28	0.26	0.222	0.2	0.0870					
0	0.083	0.16	0.22	0.25	0.279	0.29	0.297	0.3	0.3	0.3	0.3	0.3	0.297	0.292	0.28	0.25	0.215	0.2	0.0834					
0	0.08	0.15	0.21	0.25	0.275	0.29	0.296	0.3	0.3	0.3	0.3	0.3	0.296	0.289	0.28	0.25	0.209	0.2	0.0801					
0	0.077	0.15	0.2	0.24	0.271	0.29	0.295	0.3	0.3	0.3	0.3	0.3	0.295	0.287	0.27	0.24	0.203	0.1	0.0772					
0	0.075	0.14	0.2	0.24	0.267	0.28	0.293	0.3	0.3	0.3	0.3	0.3	0.293	0.284	0.27	0.24	0.198	0.1	0.0746					
0	0.072	0.14	0.19	0.23	0.264	0.28	0.291	0.3	0.3	0.3	0.3	0.3	0.291	0.281	0.26	0.23	0.193	0.1	0.0723					
0	0.07	0.13	0.19	0.23	0.26	0.28	0.29	0.3	0.3	0.3	0.3	0.3	0.29	0.279	0.26	0.23	0.189	0.1	0.0701					
0	0.068	0.13	0.18	0.23	0.256	0.28	0.288	0.29	0.3	0.3	0.3	0.3	0.288	0.276	0.26	0.23	0.185	0.1	0.0682					
0	0.066	0.13	0.18	0.22	0.253	0.27	0.286	0.29	0.3	0.3	0.3	0.3	0.286	0.273	0.25	0.22	0.181	0.1	0.0664					
0	0.065	0.12	0.18	0.22	0.249	0.27	0.284	0.29	0.29	0.29	0.3	0.284	0.271	0.25	0.22	0.177	0.1	0.0647						
0	0.063	0.12	0.17	0.21	0.246	0.27	0.282	0.29	0.29	0.29	0.3	0.282	0.268	0.25	0.21	0.173	0.1	0.0631						
0	0.062	0.12	0.17	0.21	0.243	0.27	0.28	0.29	0.29	0.29	0.3	0.28	0.265	0.24	0.21	0.17	0.1	0.0617						
0	0.06	0.12	0.17	0.21	0.24	0.26	0.278	0.29	0.29	0.29	0.3	0.278	0.263	0.24	0.21	0.167	0.1	0.0603						
0	0.059	0.11	0.16	0.2	0.237	0.26	0.276	0.29	0.29	0.29	0.3	0.276	0.26	0.24	0.2	0.164	0.1	0.0591						
0	0.058	0.11	0.16	0.2	0.234	0.26	0.274	0.28	0.29	0.29	0.3	0.274	0.258	0.23	0.2	0.161	0.1	0.0579						
0	0.057	0.11	0.16	0.2	0.231	0.26	0.272	0.28	0.29	0.29	0.3	0.272	0.255	0.23	0.2	0.158	0.1	0.0568						
0	0.056	0.11	0.16	0.2	0.228	0.25	0.27	0.28	0.29	0.29	0.3	0.27	0.253	0.23	0.2	0.156	0.1	0.0557						
0	0.055	0.11	0.15	0.19	0.226	0.25	0.267	0.28	0.28	0.28	0.3	0.267	0.25	0.23	0.19	0.153	0.1	0.0547						
0	0.054	0.1	0.15	0.19	0.223	0.25	0.265	0.28	0.28	0.28	0.3	0.265	0.248	0.22	0.19	0.151	0.1	0.053						
0	0.053	0.1	0.15	0.19	0.22	0.25	0.263	0.27	0.28	0.28	0.3	0.263	0.245	0.22	0.19	0.149	0.1	0.052						
0	0.052	0.1	0.15	0.19	0.218	0.24	0.261	0.27	0.28	0.28	0.3	0.261	0.243	0.22	0.19	0.147	0.1	0.0520						
0	0.051	0.1	0.14	0.18	0.215	0.24	0.259	0.27	0.28	0.28	0.3	0.259	0.24	0.22	0.18	0.145	0.1	0.0512						
0	0.05	0.1	0.14	0.18	0.213	0.24	0.256	0.27	0.27	0.27	0.3	0.256	0.238	0.21	0.18	0.143	0.1	0.0504						
0	0.05	0.1	0.14	0.18	0.211	0.24	0.254	0.27	0.27	0.27	0.3	0.254	0.236	0.21	0.18	0.141	0.1	0.0496						
0	0.049	0.1	0.14	0.18	0.208	0.23	0.252	0.26	0.27	0.27	0.3	0.252	0.234	0.21	0.18	0.139	0.1	0.0489						
0	0.048	0.09	0.14	0.17	0.206	0.23	0.25	0.26	0.27	0.27	0.3	0.25	0.231	0.21	0.17	0.137	0.1	0.0482						
0	0.048	0.09	0.14	0.17	0.204	0.23	0.248	0.26	0.27	0.27	0.3	0.248	0.229	0.2	0.17	0.135	0.1	0.047						
0	0.047	0.09	0.13	0.17	0.202	0.23	0.246	0.26	0.26	0.26	0.3	0.246	0.227	0.2	0.17	0.134	0.1	0.0469						
0	0.046	0.09	0.13	0.17	0.20	0.22	0.243	0.26	0.26	0.26	0.3	0.243	0.225	0.2	0.17	0.132	0.1	0.0463						
0	0.046	0.09	0.13	0.17	0.198	0.22	0.241	0.25	0.26	0.26	0.3	0.241	0.223	0.2	0.17	0.131	0.1	0.0457						
0	0.045	0.09	0.13	0.17	0.196	0.22	0.239	0.25	0.26	0.26	0.3	0.239	0.22	0.2	0.17	0.129	0.1	0.0451						
0	0.045	0.09	0.13	0.16	0.194	0.22	0.237	0.25	0.26	0.26	0.2	0.237	0.218	0.19	0.16	0.128	0.1	0.0444						
0	0.044	0.09	0.13	0.16	0.192	0.22	0.235	0.25	0.25	0.25	0.2	0.235	0.216	0.19	0.16	0.126	0.1	0.0440						
0	0.044	0.09	0.12	0.16	0.19	0.21	0.233	0.25	0.25	0.25	0.2	0.233	0.214	0.19	0.16	0.125	0.1	0.0435						
0	0.043	0.08	0.12	0.16	0.188	0.21	0.231	0.24	0.25	0.25	0.2	0.231	0.212	0.19	0.16	0.123	0.1	0.0430						

45	0	0.043	0.08	0.12	0.16	0.19	0.21	0.2	0.24	0.25	0.25	0.239	0.23	0.208	0.18	0.15	0.12	0.08	0.043
46	0	0.042	0.08	0.12	0.15	0.18	0.21	0.2	0.24	0.25	0.25	0.237	0.22	0.206	0.18	0.15	0.12	0.08	0.042
47	0	0.042	0.08	0.12	0.15	0.18	0.21	0.2	0.24	0.24	0.24	0.237	0.22	0.206	0.18	0.15	0.12	0.08	0.042
48	0	0.041	0.08	0.12	0.15	0.18	0.2	0.2	0.23	0.24	0.24	0.235	0.22	0.204	0.18	0.15	0.12	0.08	0.041
49	0	0.041	0.08	0.12	0.15	0.18	0.2	0.2	0.23	0.24	0.24	0.233	0.22	0.202	0.18	0.15	0.12	0.08	0.041
50	0	0.04	0.08	0.12	0.15	0.18	0.2	0.2	0.23	0.24	0.24	0.231	0.22	0.201	0.18	0.15	0.12	0.08	0.04
51	0	0.04	0.08	0.11	0.15	0.18	0.2	0.2	0.23	0.23	0.23	0.229	0.22	0.199	0.18	0.15	0.11	0.08	0.04
52	0	0.039	0.08	0.11	0.15	0.17	0.2	0.2	0.23	0.23	0.23	0.227	0.21	0.197	0.17	0.15	0.11	0.08	0.039
53	0	0.039	0.08	0.11	0.14	0.17	0.19	0.2	0.22	0.23	0.23	0.225	0.21	0.195	0.17	0.14	0.11	0.08	0.039
54	0	0.039	0.08	0.11	0.14	0.17	0.19	0.2	0.22	0.23	0.23	0.223	0.21	0.193	0.17	0.14	0.11	0.08	0.039
55	0	0.038	0.08	0.11	0.14	0.17	0.19	0.2	0.22	0.23	0.23	0.221	0.21	0.191	0.17	0.14	0.11	0.08	0.038
56	0	0.038	0.07	0.11	0.14	0.17	0.19	0.2	0.22	0.22	0.22	0.219	0.21	0.19	0.17	0.14	0.11	0.07	0.038
57	0	0.037	0.07	0.11	0.14	0.17	0.19	0.2	0.22	0.22	0.22	0.217	0.2	0.188	0.17	0.14	0.11	0.07	0.037
58	0	0.037	0.07	0.11	0.14	0.16	0.19	0.2	0.21	0.22	0.22	0.215	0.2	0.186	0.16	0.14	0.11	0.07	0.037
59	0	0.037	0.07	0.11	0.14	0.16	0.18	0.2	0.21	0.22	0.22	0.213	0.2	0.184	0.16	0.14	0.11	0.07	0.037
60	0	0.036	0.07	0.1	0.13	0.16	0.18	0.2	0.21	0.22	0.22	0.211	0.2	0.183	0.16	0.13	0.1	0.07	0.036
61	0	0.036	0.07	0.1	0.13	0.16	0.18	0.2	0.21	0.21	0.21	0.209	0.2	0.181	0.16	0.13	0.1	0.07	0.036
62	0	0.036	0.07	0.1	0.13	0.16	0.18	0.2	0.21	0.21	0.21	0.207	0.2	0.179	0.16	0.13	0.1	0.07	0.036
63	0	0.035	0.07	0.1	0.13	0.16	0.18	0.2	0.21	0.21	0.21	0.205	0.19	0.178	0.16	0.13	0.1	0.07	0.035
64	0	0.035	0.07	0.1	0.13	0.15	0.18	0.2	0.21	0.21	0.21	0.203	0.19	0.176	0.15	0.13	0.1	0.07	0.035
65	0	0.034	0.07	0.1	0.13	0.15	0.17	0.2	0.2	0.21	0.21	0.201	0.19	0.174	0.15	0.13	0.1	0.07	0.034
66	0	0.034	0.07	0.1	0.13	0.15	0.17	0.2	0.2	0.2	0.2	0.199	0.19	0.173	0.15	0.13	0.1	0.07	0.034
67	0	0.034	0.07	0.1	0.13	0.15	0.17	0.2	0.2	0.2	0.2	0.198	0.19	0.171	0.15	0.13	0.1	0.07	0.034
68	0	0.033	0.07	0.1	0.12	0.15	0.17	0.2	0.2	0.2	0.2	0.196	0.19	0.169	0.15	0.12	0.1	0.07	0.033
69	0	0.033	0.07	0.1	0.12	0.15	0.17	0.2	0.19	0.2	0.2	0.194	0.18	0.168	0.15	0.12	0.1	0.07	0.033
70	0	0.033	0.06	0.09	0.12	0.15	0.17	0.2	0.19	0.2	0.2	0.192	0.18	0.166	0.15	0.12	0.09	0.06	0.033
71	0	0.033	0.06	0.09	0.12	0.14	0.16	0.2	0.19	0.2	0.2	0.19	0.18	0.165	0.14	0.12	0.09	0.06	0.033
72	0	0.032	0.06	0.09	0.12	0.14	0.16	0.2	0.19	0.19	0.19	0.189	0.18	0.163	0.14	0.12	0.09	0.06	0.032
73	0	0.032	0.06	0.09	0.12	0.14	0.16	0.2	0.19	0.19	0.19	0.187	0.18	0.162	0.14	0.12	0.09	0.06	0.032
74	0	0.032	0.06	0.09	0.12	0.14	0.16	0.2	0.19	0.19	0.19	0.185	0.18	0.16	0.14	0.12	0.09	0.06	0.032
75	0	0.031	0.06	0.09	0.12	0.14	0.16	0.2	0.18	0.19	0.19	0.184	0.17	0.159	0.14	0.12	0.09	0.06	0.031
76	0	0.031	0.06	0.09	0.12	0.14	0.16	0.2	0.18	0.19	0.19	0.182	0.17	0.157	0.14	0.12	0.09	0.06	0.031
77	0	0.031	0.06	0.09	0.11	0.14	0.16	0.2	0.18	0.19	0.19	0.18	0.17	0.156	0.14	0.11	0.09	0.06	0.031
78	0	0.03	0.06	0.09	0.11	0.14	0.15	0.2	0.18	0.18	0.18	0.179	0.17	0.154	0.14	0.11	0.09	0.06	0.03
79	0	0.03	0.06	0.09	0.11	0.13	0.15	0.2	0.18	0.18	0.18	0.177	0.17	0.153	0.13	0.11	0.09	0.06	0.03
80	0	0.03	0.06	0.09	0.11	0.13	0.15	0.2	0.18	0.18	0.18	0.175	0.17	0.151	0.13	0.11	0.09	0.06	0.03
81	0	0.03	0.06	0.09	0.11	0.13	0.15	0.2	0.17	0.18	0.18	0.174	0.16	0.15	0.13	0.11	0.09	0.06	0.03
82	0	0.029	0.06	0.08	0.11	0.13	0.15	0.2	0.17	0.18	0.18	0.172	0.16	0.149	0.13	0.11	0.08	0.06	0.029
83	0	0.029	0.06	0.08	0.11	0.13	0.15	0.2	0.17	0.18	0.18	0.17	0.16	0.147	0.13	0.11	0.08	0.06	0.029
84	0	0.029	0.06	0.08	0.11	0.13	0.15	0.2	0.17	0.17	0.17	0.169	0.16	0.146	0.13	0.11	0.08	0.06	0.029
85	0	0.028	0.06	0.08	0.11	0.13	0.14	0.2	0.17	0.17	0.17	0.167	0.16	0.145	0.13	0.11	0.08	0.06	0.028
86	0	0.028	0.06	0.08	0.11	0.13	0.14	0.2	0.17	0.17	0.17	0.166	0.16	0.143	0.13	0.11	0.08	0.06	0.028
87	0	0.028	0.06	0.08	0.1	0.12	0.14	0.2	0.16	0.17	0.17	0.164	0.16	0.142	0.12	0.1	0.08	0.06	0.028
88	0	0.028	0.05	0.08	0.1	0.12	0.14	0.2	0.16	0.17	0.17	0.163	0.15	0.141	0.12	0.1	0.08	0.05	0.028
89	0	0.027	0.05	0.08	0.1	0.12	0.14	0.2	0.16	0.16	0.16	0.161	0.15	0.139	0.12	0.1	0.08	0.05	0.027
90	0	0.027	0.05	0.08	0.1	0.12	0.14	0.2	0.16	0.16	0.16	0.16	0.15	0.138	0.12	0.1	0.08	0.05	0.027

91	0	0.03	0.05	0.078	0.1	0.12	0.14	0.15	0.16	0.16	0.16	0.16	0.15	0.14	0.12	0.1	0.078	0.05	0.02
92	0	0.03	0.05	0.077	0.1	0.119	0.14	0.15	0.16	0.16	0.16	0.16	0.15	0.14	0.12	0.1	0.077	0.05	0.02
93	0	0.03	0.05	0.076	0.1	0.118	0.13	0.15	0.16	0.16	0.16	0.16	0.15	0.13	0.12	0.1	0.076	0.05	0.02
94	0	0.03	0.05	0.076	0.1	0.117	0.13	0.15	0.15	0.16	0.16	0.15	0.15	0.13	0.12	0.1	0.076	0.05	0.02
95	0	0.03	0.05	0.075	0.1	0.116	0.13	0.14	0.15	0.16	0.16	0.15	0.14	0.13	0.12	0.1	0.075	0.05	0.02
96	0	0.03	0.05	0.074	0.1	0.115	0.13	0.14	0.15	0.16	0.16	0.15	0.14	0.13	0.11	0.1	0.074	0.05	0.02
97	0	0.03	0.05	0.074	0.09	0.114	0.13	0.14	0.15	0.15	0.15	0.15	0.14	0.13	0.11	0.1	0.074	0.05	0.02
98	0	0.03	0.05	0.073	0.09	0.113	0.13	0.14	0.15	0.15	0.15	0.15	0.14	0.13	0.11	0.1	0.073	0.05	0.02
99	0	0.02	0.05	0.072	0.09	0.112	0.13	0.14	0.15	0.15	0.15	0.15	0.14	0.13	0.11	0.1	0.072	0.05	0.02
00	0	0.02	0.05	0.071	0.09	0.11	0.13	0.14	0.15	0.15	0.15	0.15	0.14	0.13	0.11	0.1	0.071	0.05	0.02
01	0	0.02	0.05	0.071	0.09	0.109	0.12	0.14	0.14	0.15	0.15	0.14	0.14	0.12	0.11	0.1	0.071	0.05	0.02
02	0	0.02	0.05	0.07	0.09	0.108	0.12	0.13	0.14	0.15	0.15	0.14	0.13	0.12	0.11	0.1	0.07	0.05	0.02
03	0	0.02	0.05	0.07	0.09	0.107	0.12	0.13	0.14	0.15	0.15	0.14	0.13	0.12	0.11	0.1	0.07	0.05	0.02
04	0	0.02	0.05	0.069	0.09	0.106	0.12	0.13	0.14	0.14	0.14	0.14	0.13	0.12	0.11	0.1	0.069	0.05	0.02
05	0	0.02	0.05	0.068	0.09	0.105	0.12	0.13	0.14	0.14	0.14	0.14	0.13	0.12	0.11	0.1	0.068	0.05	0.02
06	0	0.02	0.05	0.068	0.09	0.104	0.12	0.13	0.14	0.14	0.14	0.14	0.13	0.12	0.1	0.1	0.068	0.05	0.02
07	0	0.02	0.05	0.067	0.09	0.104	0.12	0.13	0.14	0.14	0.14	0.14	0.13	0.12	0.1	0.1	0.067	0.05	0.02
08	0	0.02	0.05	0.066	0.09	0.103	0.12	0.13	0.14	0.14	0.14	0.14	0.13	0.12	0.1	0.1	0.066	0.05	0.02
09	0	0.02	0.04	0.066	0.08	0.102	0.12	0.13	0.13	0.14	0.14	0.13	0.13	0.12	0.1	0.1	0.066	0.04	0.02
10	0	0.02	0.04	0.065	0.08	0.101	0.11	0.13	0.13	0.14	0.14	0.13	0.13	0.11	0.1	0.1	0.065	0.04	0.02
11	0	0.02	0.04	0.065	0.08	0.1	0.11	0.12	0.13	0.14	0.14	0.13	0.12	0.11	0.1	0.1	0.065	0.04	0.02
12	0	0.02	0.04	0.064	0.08	0.099	0.11	0.12	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.1	0.064	0.04	0.02
13	0	0.02	0.04	0.063	0.08	0.098	0.11	0.12	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.1	0.063	0.04	0.02
14	0	0.02	0.04	0.063	0.08	0.097	0.11	0.12	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.1	0.063	0.04	0.02
15	0	0.02	0.04	0.062	0.08	0.096	0.11	0.12	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.1	0.062	0.04	0.02
16	0	0.02	0.04	0.062	0.08	0.095	0.11	0.12	0.13	0.13	0.13	0.13	0.12	0.11	0.1	0.1	0.062	0.04	0.02
17	0	0.02	0.04	0.061	0.08	0.094	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.11	0.09	0.1	0.061	0.04	0.02
18	0	0.02	0.04	0.06	0.08	0.093	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.11	0.09	0.1	0.06	0.04	0.02
19	0	0.02	0.04	0.06	0.08	0.093	0.11	0.12	0.12	0.13	0.13	0.12	0.12	0.11	0.09	0.1	0.06	0.04	0.02
20	0	0.02	0.04	0.059	0.08	0.092	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.059	0.04	0.02
21	0	0.02	0.04	0.059	0.08	0.091	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.059	0.04	0.02
22	0	0.02	0.04	0.058	0.08	0.09	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.058	0.04	0.02
23	0	0.02	0.04	0.058	0.07	0.089	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.058	0.04	0.02
24	0	0.02	0.04	0.057	0.07	0.088	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.057	0.04	0.02
25	0	0.02	0.04	0.057	0.07	0.088	0.1	0.11	0.12	0.12	0.12	0.12	0.11	0.1	0.09	0.1	0.057	0.04	0.02
26	0	0.02	0.04	0.056	0.07	0.087	0.1	0.11	0.11	0.12	0.12	0.11	0.11	0.1	0.09	0.1	0.056	0.04	0.01

7	0	0.019	0.04	0.06	0.1	0.1	0.1	0.107	0.113	0.116	0.116	0.11	0.1069	0.1	0.09	0.1	0.056	0.04	0.0
8	0	0.019	0.04	0.06	0.1	0.1	0.1	0.106	0.112	0.115	0.115	0.11	0.106	0.1	0.09	0.1	0.055	0.04	0.
9	0	0.019	0.04	0.05	0.1	0.1	0.1	0.105	0.111	0.114	0.114	0.11	0.105	0.1	0.08	0.1	0.055	0.04	0.0
0	0	0.019	0.04	0.05	0.1	0.1	0.1	0.104	0.11	0.113	0.113	0.11	0.104	0.1	0.08	0.1	0.054	0.04	0.0
1	0	0.019	0.04	0.05	0.1	0.1	0.09	0.103	0.109	0.112	0.112	0.11	0.103	0.09	0.08	0.1	0.054	0.04	0.0
2	0	0.018	0.04	0.05	0.1	0.1	0.09	0.102	0.108	0.111	0.111	0.11	0.1021	0.09	0.08	0.1	0.053	0.04	0.0
3	0	0.018	0.04	0.05	0.1	0.1	0.09	0.101	0.107	0.11	0.11	0.11	0.1011	0.09	0.08	0.1	0.053	0.04	0.0
4	0	0.018	0.04	0.05	0.1	0.1	0.09	0.1	0.106	0.109	0.109	0.11	0.1002	0.09	0.08	0.1	0.052	0.04	0.
5	0	0.018	0.04	0.05	0.1	0.1	0.09	0.099	0.105	0.108	0.108	0.11	0.0993	0.09	0.08	0.1	0.052	0.04	0.0
6	0	0.018	0.03	0.05	0.1	0.1	0.09	0.098	0.104	0.107	0.107	0.1	0.0983	0.09	0.08	0.1	0.051	0.03	0.0
7	0	0.018	0.03	0.05	0.1	0.1	0.09	0.097	0.103	0.106	0.106	0.1	0.0974	0.09	0.08	0.1	0.051	0.03	0.0
8	0	0.017	0.03	0.05	0.1	0.1	0.09	0.097	0.102	0.105	0.105	0.1	0.0965	0.09	0.08	0.1	0.05	0.03	0.0
9	0	0.017	0.03	0.05	0.1	0.1	0.09	0.096	0.101	0.104	0.104	0.1	0.0956	0.09	0.08	0.1	0.05	0.03	0.0
0	0	0.017	0.03	0.05	0.1	0.1	0.09	0.095	0.1	0.103	0.103	0.1	0.0948	0.09	0.08	0.1	0.049	0.03	0.
1	0	0.017	0.03	0.05	0.1	0.1	0.09	0.094	0.099	0.102	0.102	0.1	0.0939	0.09	0.08	0.1	0.049	0.03	0.0
2	0	0.017	0.03	0.05	0.1	0.1	0.09	0.093	0.098	0.101	0.101	0.1	0.093	0.09	0.07	0.1	0.048	0.03	0.0
3	0	0.017	0.03	0.05	0.1	0.1	0.08	0.092	0.098	0.1	0.1	0.1	0.0921	0.08	0.07	0.1	0.048	0.03	0.0
4	0	0.016	0.03	0.05	0.1	0.1	0.08	0.091	0.097	0.099	0.099	0.1	0.0913	0.08	0.07	0.1	0.047	0.03	0.0
5	0	0.016	0.03	0.05	0.1	0.1	0.08	0.09	0.096	0.098	0.098	0.1	0.0904	0.08	0.07	0.1	0.047	0.03	0.0
6	0	0.016	0.03	0.05	0.1	0.1	0.08	0.09	0.095	0.098	0.098	0.09	0.0896	0.08	0.07	0.1	0.047	0.03	0.0
7	0	0.016	0.03	0.05	0.1	0.1	0.08	0.089	0.094	0.097	0.097	0.09	0.0888	0.08	0.07	0.1	0.046	0.03	0.
8	0	0.016	0.03	0.05	0.1	0.1	0.08	0.088	0.093	0.096	0.096	0.09	0.0879	0.08	0.07	0.1	0.046	0.03	0.0
9	0	0.016	0.03	0.05	0.1	0.1	0.08	0.087	0.092	0.095	0.095	0.09	0.0871	0.08	0.07	0.1	0.045	0.03	0.0
0	0	0.016	0.03	0.04	0.1	0.1	0.08	0.086	0.091	0.094	0.094	0.09	0.0863	0.08	0.07	0.1	0.045	0.03	0.0
1	0	0.015	0.03	0.04	0.1	0.1	0.08	0.086	0.091	0.093	0.093	0.09	0.0855	0.08	0.07	0.1	0.044	0.03	0.0
2	0	0.015	0.03	0.04	0.1	0.1	0.08	0.085	0.09	0.092	0.092	0.09	0.0847	0.08	0.07	0.1	0.044	0.03	0.0
3	0	0.015	0.03	0.04	0.1	0.1	0.08	0.084	0.089	0.091	0.091	0.09	0.0839	0.08	0.07	0.1	0.044	0.03	0.0
4	0	0.015	0.03	0.04	0.1	0.1	0.08	0.083	0.088	0.091	0.091	0.09	0.0832	0.08	0.07	0.1	0.043	0.03	0.0
5	0	0.015	0.03	0.04	0.1	0.1	0.08	0.082	0.087	0.09	0.09	0.09	0.0824	0.08	0.07	0.1	0.043	0.03	0.0
6	0	0.015	0.03	0.04	0.1	0.1	0.07	0.082	0.086	0.089	0.089	0.09	0.0816	0.07	0.07	0.1	0.042	0.03	0.0
7	0	0.015	0.03	0.04	0.1	0.1	0.07	0.081	0.086	0.088	0.088	0.09	0.0809	0.07	0.06	0.1	0.042	0.03	0.0
8	0	0.014	0.03	0.04	0.1	0.1	0.07	0.08	0.085	0.087	0.087	0.08	0.0801	0.07	0.06	0.1	0.042	0.03	0.0
9	0	0.014	0.03	0.04	0.1	0.1	0.07	0.079	0.084	0.086	0.086	0.08	0.0794	0.07	0.06	0.1	0.041	0.03	0.0
0	0	0.014	0.03	0.04	0.1	0.1	0.07	0.079	0.083	0.086	0.086	0.08	0.0786	0.07	0.06	0.1	0.041	0.03	0.0
1	0	0.014	0.03	0.04	0.1	0.1	0.07	0.078	0.082	0.085	0.085	0.08	0.0779	0.07	0.06	0.1	0.04	0.03	0.
2	0	0.014	0.03	0.04	0.1	0.1	0.07	0.077	0.082	0.084	0.084	0.08	0.0772	0.07	0.06	0.1	0.04	0.03	0.0
3	0	0.014	0.03	0.04	0.1	0.1	0.07	0.076	0.081	0.083	0.083	0.08	0.0765	0.07	0.06	0.1	0.04	0.03	0.0
4	0	0.014	0.03	0.04	0.1	0.1	0.07	0.076	0.08	0.082	0.082	0.08	0.0758	0.07	0.06	0.1	0.039	0.03	0.0
5	0	0.013	0.03	0.04	0.1	0.1	0.07	0.075	0.079	0.082	0.082	0.08	0.0751	0.07	0.06	0.1	0.039	0.03	0.0
6	0	0.013	0.03	0.04	0	0.1	0.07	0.074	0.079	0.081	0.081	0.08	0.0744	0.07	0.06	0	0.039	0.03	0.0
7	0	0.013	0.03	0.04	0	0.1	0.07	0.074	0.078	0.08	0.08	0.08	0.0737	0.07	0.06	0	0.038	0.03	0.0
8	0	0.013	0.03	0.04	0	0.1	0.07	0.073	0.077	0.079	0.079	0.08	0.073	0.07	0.06	0	0.038	0.03	0.0
9	0	0.013	0.03	0.04	0	0.1	0.07	0.072	0.077	0.079	0.079	0.08	0.0723	0.07	0.06	0	0.038	0.03	0.
0	0	0.013	0.03	0.04	0	0.1	0.07	0.072	0.076	0.078	0.078	0.08	0.0717	0.07	0.06	0	0.037	0.03	0.0
1	0	0.013	0.03	0.04	0	0.1	0.06	0.071	0.075	0.077	0.077	0.08	0.071	0.06	0.06	0	0.037	0.03	0.0



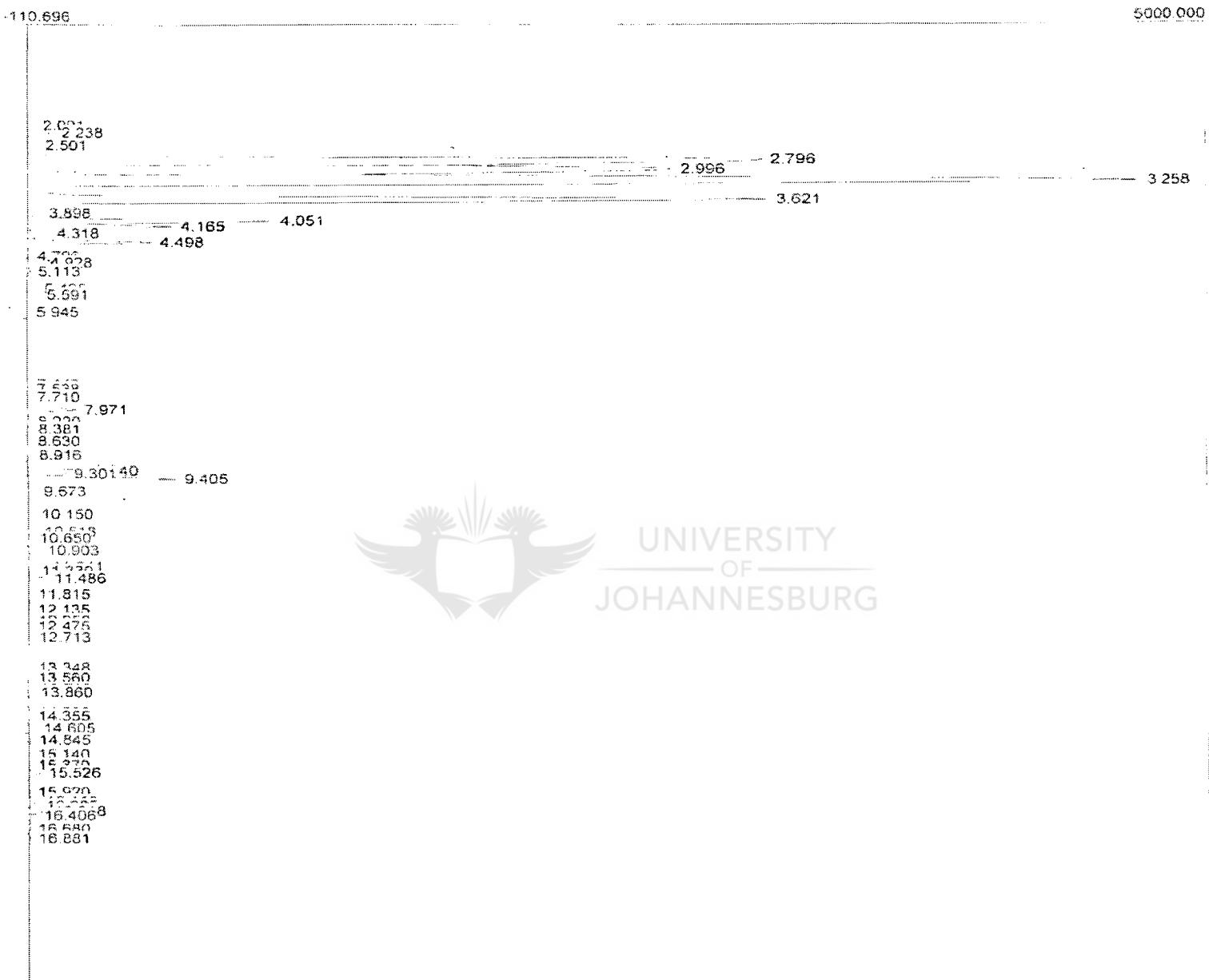


255	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.04	0.04	0.04	0.03	0.03	0.03	0.03	0.02	0.017	0.012	0.0		
256	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.04	0.04	0.04	0.03	0.03	0.03	0.03	0.02	0.017	0.011	0.0		
257	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.017	0.011	0.0		
258	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.016	0.011	0.0		
259	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.016	0.011	0.0		
260	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.016	0.011	0.0	
261	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.016	0.011	0.0	
262	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.016	0.011	0.0	
263	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.016	0.011	0.0	
264	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.016	0.011	0.0	
265	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
266	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
267	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
268	0	0.01	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
269	0	0.01	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
270	0	0.01	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
271	0	0.01	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.015	0.01	0.0	
272	0	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.014	0.01	0.0	
273	0	0	0.01	0.01	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.014	0.01	0.0	
274	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.014	0.01	0.0
275	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.014	0.01	0.0
276	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.014	0.009	0.0
277	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.014	0.009	0.0
278	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.014	0.009	0.0
279	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.013	0.009	0.0
280	0	0	0.01	0.01	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.013	0.009	0.0



## Appendix C: Typical chromatograms for the Eucalyptus oil extract.

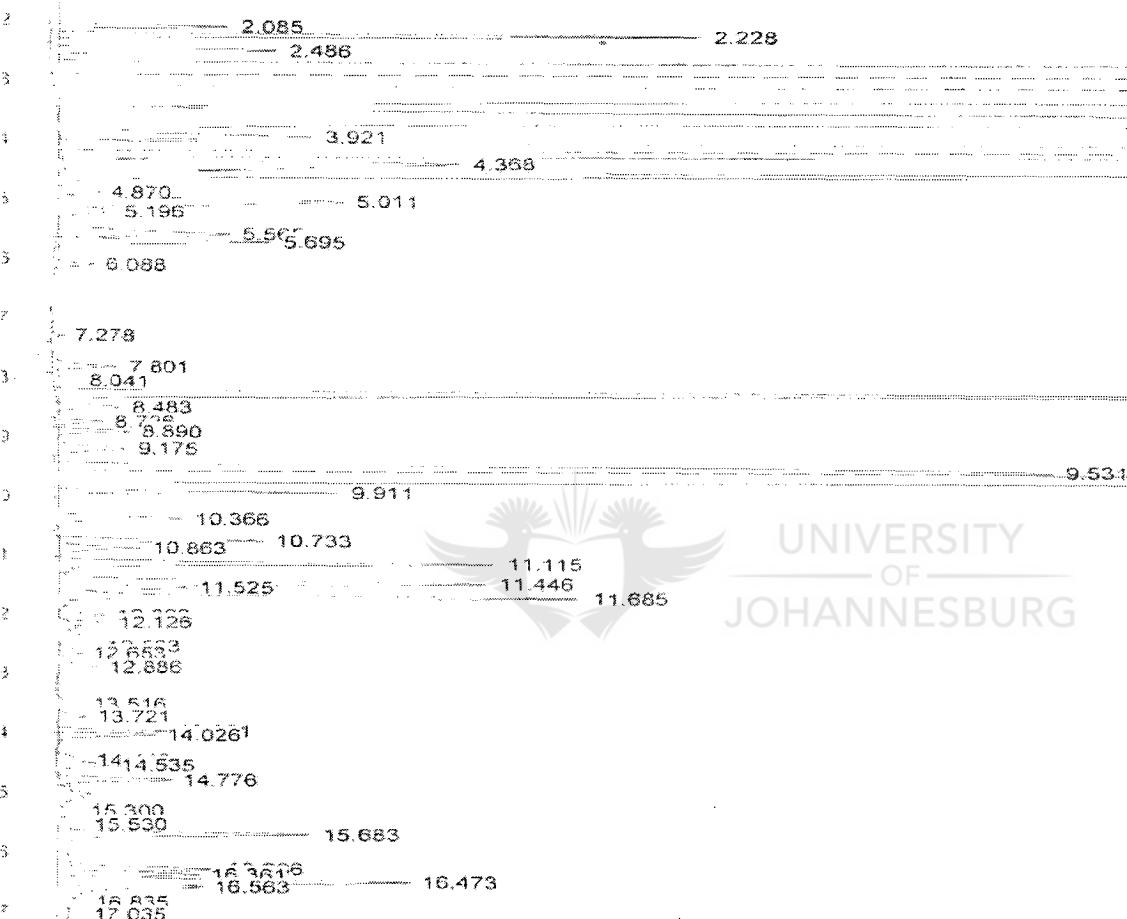
Lab name: SRI Instruments  
Analysis date: 04/26/2007 11:05:58  
Method: Syringe Injection  
Description: FID  
Carrier: HELIUM AT 5 PSI  
Data file: Eucalyptus 1.CHR ()  
Sample: Eucalyptus 1



**Figure C1: Chromatogram of Eucalyptus oil extract at 100kPa**

\* Lab name: SRI Instruments  
Analysis date: 04/26/2007 11:37:22  
Method: Syringe Injection  
Description: FID  
Carrier: HELIUM AT 5 PSI  
Data file: Eucalyptus 2.chr ()  
Sample: Eucalyptus 1

-10.134



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Figure C2: Chromatogram of Eucalyptus oil extract at 1500kPa