



Seasonal variability of Nitrous Oxide (N_2O) in Rwanda

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Declaration

I declare that this Dissertation contains my own work except where specifically acknowledged.

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Signature.....

Date.....

Acknowledgement

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Abstract

N_2O is the third most important greenhouse gas after carbon dioxide (CO_2) and methane (CH_4). It contributes to global warming and ozone layer depletion though not given too much attention compared to CO_2 and CH_4 . It is necessary to assess the variability of N_2O in Rwanda in order to get its status and how N_2O concentration varies with seasons.

N_2O data were measured using the mid infrared analyzer, also known as Picaro G5105. The data used have been recorded from Rwanda Climate observatory, Mugogo station during the year 2017, starting from January up to December. Up on analysis of N_2O data, it has been found that the concentration of Nitrous oxide increased during the year 2017. The N_2O concentration passed from the 329.79 *ppb* during the January to February season to 330.15 *ppb* during the October to December season. The mean annual concentration for Nitrous oxide was 329.5 *ppb*. That mean value of N_2O concentration shows that the concentration of N_2O continues to increase, because the N_2O concentration previously reported ranged from 320 to 324 *ppb* in the atmosphere.

The seasonal variation of N_2O showed that the [March-April-May \(MAM\)](#) season had the lowest mean concentration of 329.08 *ppb*. The sharp decrease is probably due to lack of coupling between the soil temperature and rainfall received by the soil. The next lowest seasonal concentration of N_2O was found during the [June-July-August-September \(JJAS\)](#) season. [JJAS](#) season's concentration was 329.30 *ppb*. Though the concentration was still below the one found during the [January-February \(JF\)](#) season, it had increased compared to [MAM](#) season. The reason behind that increase may have been the addition of nitrogen fertilizers and manure to soil, because it was a growing season. But also because N_2O is a long lived species, the contribution from remote locations may be taken into account.

The highest seasonal concentration is found in the [October-November-December \(OND\)](#) season with a value of 330.15 *ppb*. The reason is the rainfall received, and temperature which is higher compared to the one experienced during the [MAM](#) season.

The seasonal patterns of the N_2O for Rwanda and Australia exhibit almost the same behaviour. The correlation coefficient between Nitrous oxide concentration and temperature, and humidity as well, yielded the values of 0.1085 and -0.1325 respectively for temperature and for humidity. The values suggested that the correlation is negative with RH and positive with T although it is still not significant and is therefore weak.

For better elucidation of the seasonal dependence of N_2O concentration variation, data for more than one year should be used for future research. The use of inventories estimation shall help in quantifying the N_2O emissions by sectors and non-in-country emissions.

Key Words

Greenhouse gas

Global Warming Potential

Ozone Depletion Potential

Nitrous oxide

Denitrification

Nitrification

Nitrogen cycle

Acronyms

AGAGE Advanced Global Atmospheric Gases Experiment [3](#)

aq aqueous [12](#)

DAP Diammonium Phosphate [5](#)

DAS Data Acquisition System [21](#)

DCD Dicyandiamide [18](#)

DMPP 3,4-Dimethylpyrazole phosphate [18](#)

DNDC Denitrification and Decomposition [17](#)

FTIR Fourier Transform infrared [17](#)

GHGs Greenhouse Gases [1](#)

GWP Global Warming Potential [2](#), [6](#), [8](#), [9](#)

HFCs Hydrofluorocarbons [8](#)

Hysplit Hybrid Single Particle Lagrangian Integrated Trajectory [23](#)

IPCC Intergovernmental Panel on Climate Change [17](#)

JF January-February [iii](#), [5](#), [24](#), [27](#), [28](#), [29](#)

JJAS June-July-August-September [iii](#), [5](#), [24](#), [27](#), [28](#)

m mass [14](#)

MAM March-April-May [iii](#), [5](#), [24](#), [27](#), [28](#), [29](#)

MDGs Millenium Development Goals [5](#)

Mt Mega ton [6](#)

N Nitrogen [15](#)

NOAA National Oceanic and Atmospheric Administration [23](#)

NPK Nitrogen Phosphorus Potassium 5

ODP Ozone Depletion Potential 14

OND October-November-December iii, 5, 24, 27, 28, 29

PFCs Perfluorocarbons 8

pH potential of Hydrogen Ion 16

ppb parts per billion 9

PPU Pump Power Unit 21, 22

R correlation coefficient 24, 25, 26

RF Radiative Forcing 9

RH Humidity 6, 24, 25

STD standard deviation 24, 26

T Temperature 6, 24, 25

UNFCC United Nations Framework Convention on Climate Change 2

WFPS Water Filled Pore Spaces 15, 28

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Chapter 1

Introduction

1.1 Background

Around 1750s, with the start of industrial age, significant increases in the atmospheric concentration of several trace gases which can affect environment on regional and global scales have occurred. Nitrous oxide (N_2O) is one of the three trace gases besides carbon dioxide (CO_2) and Methane (CH_4), which are the main [Greenhouse Gases \(GHGs\)](#) and therefore contributing to global warming [52].

N_2O levels have risen approximately 15% since the preindustrial times, reaching 315ppb in 2000 [45] and $324 \pm 0.1 \text{ ppb}$ in 2011, with an increase of 5 *ppb* since 2005 [6].

International policy discussions have focused in the last two decades on non- CO_2 emissions like N_2O because they are less expensive to mitigate than CO_2 emissions [12].

Nitrous oxide is a colorless gas of slightly sweet odor and taste under ambient conditions. It was discovered by Joseph Priestley in 1772 from the reduction of Nitric oxide (NO) with iron sulfur mixtures [52]. The gas is used as anesthetic and it is commonly referred to as *laughing gas* [45].

N_2O is present in the earth's atmosphere at a trace level and its mixing ratio was of order of 324 ppb in 2010, with an increase rate of 0.75 ppb per year [28]. The concentration of Nitrous oxide has been increasing linearly over last few decades as a consequence of the introduction of N_2O into the atmosphere at a rate greater than its rate of removal by natural processes [49]. The current estimate for the lifetime of N_2O is 120 years [45]. Nitrous oxide is produced from a wide variety of natural and human sources. Natural sources are primarily bacterial decomposition of nitrogen in soils and the earth's oceans. Anthropogenic or human related sources are agricultural activities, animal manure, sewage treatment, mobile and stationary combustion of fossil fuels and nitric acid production [42]. Biomass burning also adds to the N_2O sources[50] but also smaller quantities may result from degassing of irrigation water [45].

Soils are the main sources of nitrous oxide (N_2O) in the atmosphere, via the microbial processes of nitrification and denitrifications [26].

The main removal mechanism (sink) for nitrous oxide is through photolysis and oxidation reactions in the stratosphere [6],[55].

From recent polar ice analysis and measurements, the increasing rate due to human activities was estimated to be in the range 0.5-1.2 ppbv/yr and the estimate of global N_2O emission was estimated to range widely from 10 to 17 TgN/yr[19].

Nitrous oxide is environmentally important for two reasons. First its capacity to absorb infrared radiation or its [Global Warming Potential \(GWP\)](#) is 300 times greater than that of Carbon dioxide though its mixing ratio is a thousand times less than that of CO_2 [49].

The capacity to absorb infrared radiation is known as [GWP](#) [34]. Second reason that makes N_2O of environmental concern is that when it reaches the stratosphere along with some halogen containing compounds, contributes to ozone depletion [49]. N_2O has the third largest radiative forcing of the anthropogenic gases, at $0.17\pm 0.03 Wm^2$ with an increase of 6% since 2005 [6].

Human activities contributions to atmospheric Nitrous oxide (N_2O) are now about the same as the contributions from natural systems [50].

This occurs as a result of addition reactive forms of nitrogen into the biosphere beyond the natural additions from, biological nitrogen fixation by leguminous plants or plants with other symbiotic associations with microorganisms and free living bacteria, and lightning flashes in the atmosphere.

Mainly the reactive forms come from the addition of synthetic nitrogenous fertilizers and animal manure to agricultural land as well as to the creation of new arable areas (land) from forests and grasslands, which cause the liberation of nitrogen from relatively inert forms from the soil and therefore releasing reactive forms of nitrogen into the atmosphere [49]. Because of its heavy reliance on synthetic Nitrogen fertilizers, agriculture has enhanced the processes of nitrification and denitrification, as a result of the two processes, agro-ecosystems contribute 55-65% of the global anthropogenic emissions of N_2O [26].

As required by the Kyoto agreement that countries have to submit the annual reports of anthropogenic greenhouse gases emissions to the [United Nations Framework Convention on Climate Change \(UNFCCC\)](#), some methodologies are used to estimate the emissions. The tier 1 emission factor approach is the mainly used method that estimates 1% of organic or mineral N applied to soils to be emitted as N_2O . The same approach estimates manure storage systems to emit 2% of N manure content as N_2O [48]. Whereas [34] assumed that 2% of anthropogenic N leaks off as N_2O and found that atmospheric N_2O has an increasing shape over the last 150 years.

[43] used the three-dimensional chemical transport model, Model for ozone and related chemical tracers version 4 (MOZART v4) and a Bayesian inverse method to estimate global as well as regional annual N_2O emissions for five sources sectors from 13 regions in the world.

The instrumentations and calibrations used are from [Advanced Global Atmospheric Gases Experiment \(AGAGE\)](#).

The study showed an increasing trend of N_2O emissions between 1995 and 2008. The results found by [43] for the period between 1997 and 2001 give a global total flux of [16.16-17.44] Tg N_2O -N/yr which is in agreement with the one found by [15] for the same period which is [15.1-17.8] N_2O -N/yr.

The study of [48] on *Nitrous oxide, Climate change and Agriculture* gives the estimates of nitrous oxide emissions by continent in 2010.

Africa had the 0.8Tg N_2O , America had 1.7Tg N_2O , Asia had the greatest emission of 3.0Tg N_2O Europe had 0.87Tg N_2O and Oceania with the lowest emission of 0.2Tg N_2O .

The data of N_2O for countries such as Australia and New Zealand, which are countries located in the same latitudinal range as Rwanda, show that N_2O concentration has been increasing for the 2 last decades, as shown in the [Figure 1.1.1](#) below. The figure is generated using the data from [1].

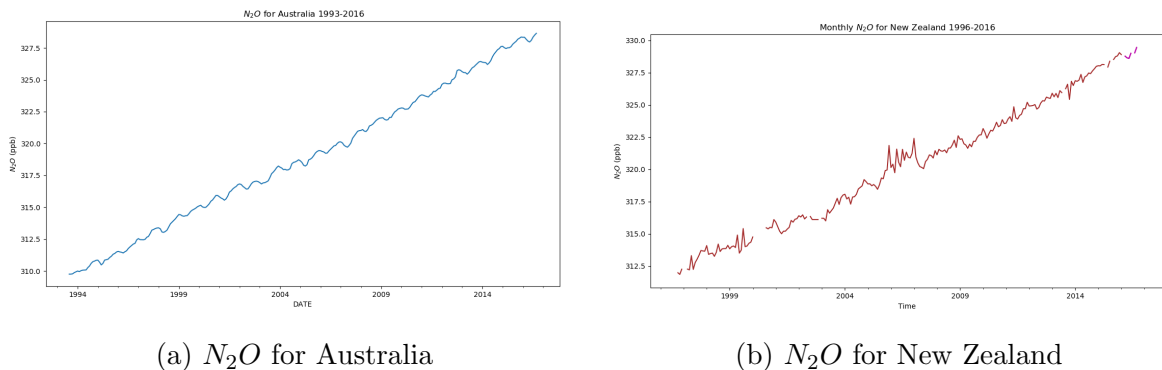


Figure 1.1.1: The N_2O concentration pattern for Australia and New Zealand during the last 2 decades, with data obtained from [1]

The concentration of N_2O in the southern hemisphere are lower during March, April and May compared to the rest of the year [25].

The [Figure 1.1.2](#) below shows the N_2O concentration pattern for the 9 months ,January to September, using the data of N_2O for Australia, recorded from Cape Grim station.

The sampling type used was "continuous", while the observation category was "air sampling observation at a stationary platform"[1].

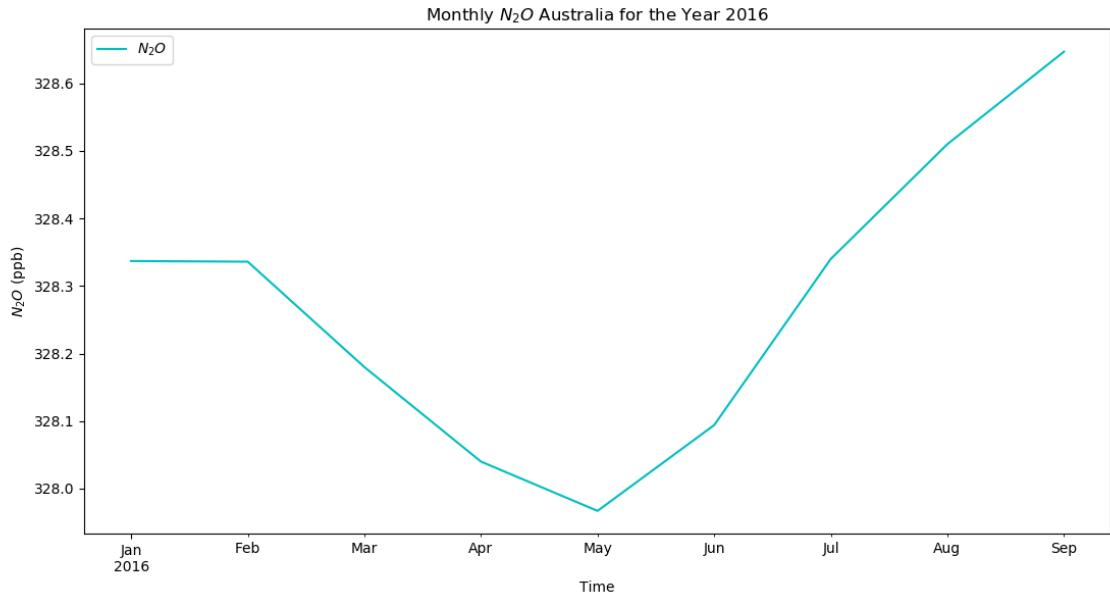


Figure 1.1.2: The N_2O concentration for nine months (January to September) of the year 2016, figure drawn using the data from [1]

From the Figure 1.1.2 above, the observation is that March to May has a decreasing shape as evidenced by [25].

The same information is presented using the bar plot as show in the Figure 1.1.3 below.

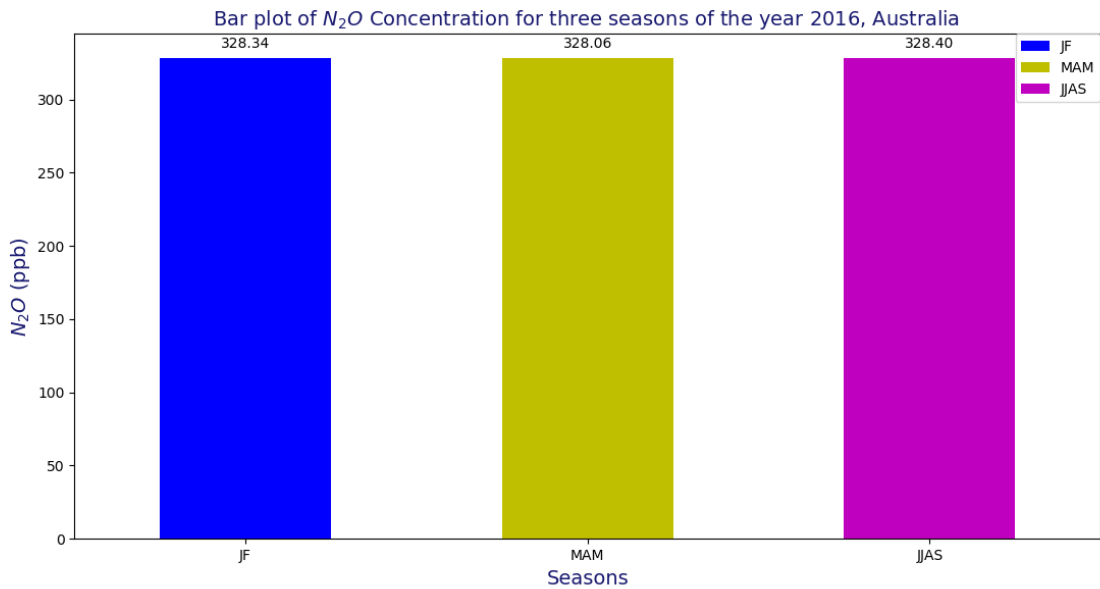


Figure 1.1.3: The bar plot of N_2O concentration for nine months (January to September, grouped in three seasons) of the year 2016, figure drawn using the data from [1]

In Rwanda we have two dry and two rainy seasons. Short dry season is the **JF** season and the long dry season is the **JJAS** season. The short rainy season is the **OND** season and the long rainy season is the **MAM** season [30].

This research aims at comparing the concentrations of N_2O for the dry and rainy seasons. The needed data are obtained through new high frequency greenhouse monitoring station [14]. The mid-infrared analyzer also known as Picarro G5105 is used to measure the N_2O concentration data to be analyzed.

1.2 Problem statement

Rwanda as many other developing countries all over the world strive to increase the production, either industrial or agricultural/farming production. The quantity of materials needed to get the desired production increases as time passes by.

In agriculture a greater amount of chemical or synthetic fertilizers and manure is needed to achieve a desirable crop production in order for governments to meet the **Millenium Development Goals (MDGs)**, such as eradication of extreme poverty and hunger. Processing of increased quantities by Industries lead to increased quantity of fossil fuels burnt to produce the needed energy.

In Rwanda synthetic chemical fertilizers are used to improve the production and assist the country to go from subsistence agriculture to modern one which is market oriented [4].

Many of synthetic fertilizers used in Rwanda are nitrogenous synthetic fertilizers such as Urea (CH_4N_2O), **Nitrogen Phosphorus Potassium (NPK)** and **Diammonium Phosphate (DAP)** [24]. The percentage of crop producing households purchasing chemical fertilizers increased from 10.8% for 2005/2006 to 28.9% for 2010/2011 and reached 36.4% for 2013/2014 [35].

The use of Nitrate and ammonium based fertilizers leads to the release of significant amounts of $N_2O - N$ in the atmosphere though their converted percentage may be small. Further conversion of nitrogenous fertilizers to $N_2O - N$ may result from the leaching of those fertilizers in groundwater[56].

Current anthropogenic emissions of $N_2O - N$ are estimated at 6.7 Tg N_2O-N year-1 of which 2.8 Tg $N_2O - N$ year-1 are from agricultural activities [52].

The temperature in Rwanda has increased by $1.4^\circ C$ since 1947, an increase which is higher than the global average[16] . The global annual average temperature increase in 2017 was $0.9^\circ C$ [33].

The rise in temperature will affect humans and animals' health by increasing the spread of vector borne diseases and therefore negatively impacting crop yield, food security and thus affecting the livelihoods of the people [16].

N_2O is a green house gas contributing to global warming and ozone depletion[40].

1.3 Interest of the Research

N_2O is a potent green house gas, its capacity to trap heat or **GWP** is 300 times than that of CO_2 on molecular basis. The major anthropogenic source of N_2O is agriculture. Agriculture is developing fast. Rwanda as a developing country has a target to continue to develop its agriculture and increase the production. The country has a goal to transform agriculture from subsistence to modern one which is market oriented, thus there is use inorganic fertilizers to achieve the target. Rwanda imported 36000 **Mega ton (Mt)** of inorganic fertilisers in 2014 and importations were supposed to increase thereafter [41] .

This research is intended to give the status of the actual mole fraction or concentration of N_2O and the picture of how N_2O varies with seasons. The outcome of this research may help decision and policy makers to devise mitigation measures to reduce the N_2O emission in the atmosphere. This research may lay the basis to further researches in Rwanda and in the entire Africa because the topics about nitrous oxide seem to be still lagging behind.

1.4 Objectives of the research

1.4.1 Main objective

The main objective of the research is to evaluate the seasonal variability of N_2O concentration in Rwanda.

1.4.2 Specific objectives

The objective of this project is to quantify the seasonal variability of N_2O concentration as one of the potent greenhouse gases causing global warming and ozone layer depletion.

The aim extends to finding the possible correlation between N_2O concentration variability with weather parameters such as **Temperature (T)** and **Humidity (RH)**.

1.5 Organization of the thesis

This thesis is composed of five chapters. The first chapter is INTRODUCTION, which is composed by the Background about nitrous oxide, problem statement, Interest of the research and objectives of the research .

The Second chapter is LITERATURE REVIEW, which includes Overview on greenhouse gases, nitrogen cycle, photochemistry and chemistry of N_2O , ozone depletion potential, drivers of N_2O soil emissions, methods used for N_2O measurement (assessments) and Mitigation measures proposed for N_2O emission reduction.

The third chapter is METHODOLOGY, where there is description of Site selection, Measurement of Nitrous oxide and data analysis. The fourth chapter is RESULTS AND INTERPRETATION and the Fifth chapter comes as CONCLUSION AND RECOMMENDATION.

Chapter 2

Literature Review

2.1 Greenhouse gases Overview

2.1.1 Definition

Greenhouse gases are gaseous constituents of the atmosphere, either natural or anthropogenic which absorb and emit radiation at specific wavelengths within the spectrum of thermal infrared radiation emitted by the earth's surface, by the atmosphere itself and by clouds [17]. The trapping of heat by greenhouse gases through the absorption of infrared radiation reflected or emitted by the earth is called the *greenhouse effect*.

It is a natural phenomenon that keeps earth's surface average temperatures around $18^{\circ}C$, which would otherwise be $-15^{\circ}C$. Therefore the phenomenon of greenhouse makes life possible on earth [51].

Besides CO_2 , N_2O and CH_4 which are the main greenhouse gases, Kyoto protocol deals with a number of entirely human-made greenhouse gases in the atmosphere such as halocarbons, bromine and chlorine containing substances. The latter gases are sulfur hexafluoride (SF_6), [Hydrofluorocarbons \(HFCs\)](#) and [Perfluorocarbons \(PFCs\)](#) [17].

2.1.2 Global warming potentials and lifetimes of major greenhouse gases

[GWP](#) is defined as the ratio of the time integrated radiative forcing from one kilogram of a well mixed Greenhouse gas relative to that of one kilogram of CO_2 over a specified time horizon. It is a useful index to compare the radiative effects of different gases relative to that of CO_2 .

GWP can be calculated by using the formula below:

$$GWP(X) = \frac{\int_0^t RF_X[X(t)]dt}{\int_0^t RF_{CO_2}[CO_2(t)]dt} \quad (2.1.1)$$

where \mathbf{t} is the time horizon (typically 20, 100, or 500 years) over which the integration is performed, where the **Radiative Forcing (RF)** (calculated change in radiation flux at the tropopause) is resulting from a unit increase in the atmospheric concentration of a given species and has units of $Wm^{-2} ppb^{-1}$, $[\mathbf{x}(\mathbf{t})]$ and $[CO_2(t)]$ are concentrations (in units of **parts per billion (ppb)**) of x and CO_2 as a function of time following instantaneous release of 1 kg of each substance[53].

The lifetimes and **GWP** of major green house gases and that of CF_4 and HFCs according to [18] is given in the **Table 2.1.1** below:

Table 2.1.1: GWP and lifetimes for major greenhouse gases

Greenhouse Gases	Lifetimes (years)	Global warming potentials (GWP)	
		Cumulative forcing over 20 years	Cumulative forcing over 100 years
CO_2	No single value can be given to CO_2	1	1
CH_4	12.4	84	28
N_2O	121.0	264	265
CF_4	50,000.0	4880	6630
$HFC - 152$	1.5	506	138

2.2 Nitrogen cycle

Below is the figure 3.2.1 explaining the nitrogen cycle:

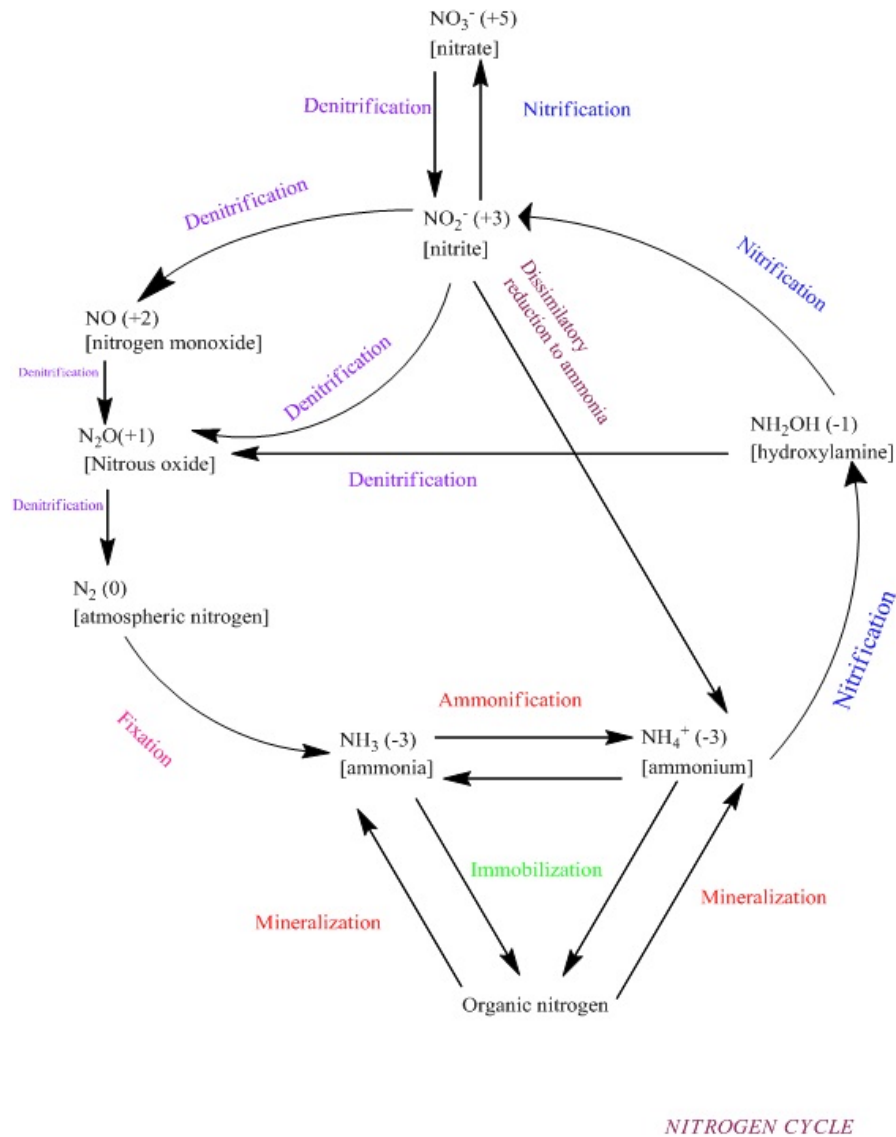
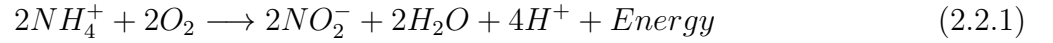


Figure 2.2.1: Nitrogen cycle with numbers in parenthesis being the oxidation states for nitrogen [47]

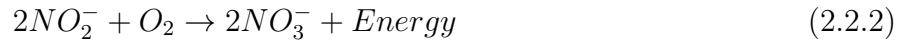
2.2.1 Nitrification

Nitrification is the aerobic oxidation of NH_4^+ to NO_3^- by chemoautotrophic bacteria. It has two stages: Nitritation, which takes place by the oxidation of NH_4^+ to NO_2^- by *Nitrosomonas* sp., *Nitrospira* sp. and *Nitrosococcus* sp.; and Nitratation in which NO_2^- is oxidized to NO_3^- by *Nitrobacter* sp., *Nitrospira* and *Nitrosococcus* sp.

Nitritation :

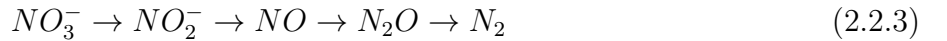


Nitratation:

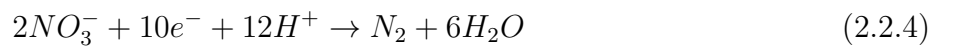


2.2.2 Denitrification

It is the reduction of NO_3^- to N_2 and the process takes place in the presence of anaerobic bacteria. Denitrification can be complete resulting in N_2 or be incomplete resulting in a fraction of Nitrogen being emitted as NO and NO_2^- .

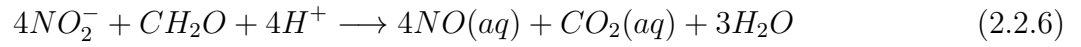
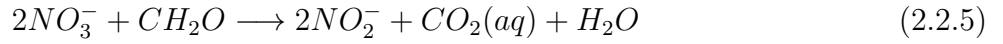


This occurs when soil is saturated and bacteria use nitrate as oxygen source [21]. The redox reaction for denitrification according to [5] is as below:



The denitrification process occurs in four steps, namely the conversion of NO_3^- to NO_2^- , NO_2^- to NO , NO to N_2O and then the conversion of N_2O to inert N_2 [20].

The detailed reaction according to [20] are the following:



,with **aqueous (aq)** meaning that there is presence of water. Some denitrifying bacteria include species in the genera of bacillus, Paracoccus and Pseudomonas. Denitrification is important for it removes fixed nitrogen, usually nitrate, from the ecosystem back to the atmosphere in a biologically inert form, usually N_2 .

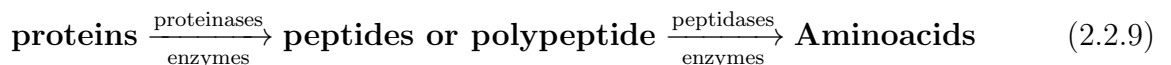
2.2.3 Ammonification

In the process of proteolysis, amino acids and proteins are synthesized from non symbiotic and symbiotic ammonia of nitrogen fixation. Animals eat plant protein and synthesize animal proteins. On death, proteins are broken down by microorganisms with the help of proteolysis enzymes. The breakdown of proteins is carried out in two stages:

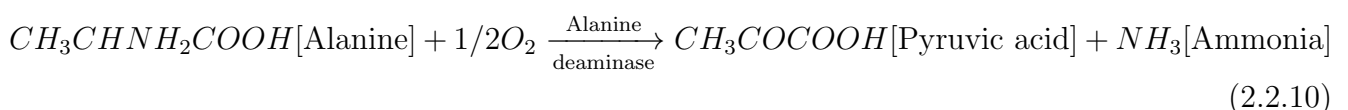
Stage 1: the proteins are converted into peptides and polypeptides by proteinases enzymes.

Stage 2: polypeptides or peptides are broken down into amino acids by peptidases enzymes [3].

The summary of proteolysis is shown below:

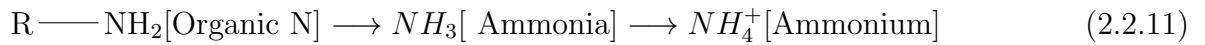


Ammonification is the process by which the amino acid released during proteolysis undergoes deamination and the NH_2 group is removed. The process is brought about by Clostridium sp., Micrococcus sp. and Proteus sp. The process structure by [3] is shown below:



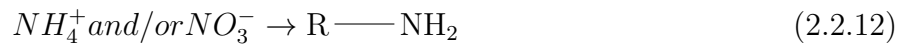
2.2.4 Mineralization

Mineralization is the process by which microbes decompose organic N from manure, Organic matter and crop residues to ammonium as illustrated by [21] below:



2.2.5 Immobilization

Immobilization is the reverse of mineralization, it is the process in which nitrates and ammonium are taken up by soil organisms and therefore become unavailable to crop.



2.3 Photochemistry and chemistry of N_2O

The only significant sink for N_2O is photolysis in the stratosphere where about 80% of it is reaching the stratosphere is destroyed in the reaction producing N_2 according to reaction below:



[44]

And about 20% is destroyed in the reaction with $O(^1D)$ produced from the equation above according to the reaction



[44]

But a small amount can form NO:



The nitric oxide produced above is the one that destroys ozone according to the reactions below :





The overall reaction for the three equations becomes



The overall lifetime of NO is too short in the troposphere for an appreciable amount to reach the stratosphere where it might cause ozone depletion. So most of stratospheric NO is derived from stratospheric N_2O and only a small amount is contributed by high altitude aircrafts [44].

2.4 Ozone depletion potential (ODP)

Ozone Depletion Potential (ODP) is widely used in quantifying the relative ozone destroying capabilities of compounds. It is defined as the time integrated global ozone depletion induced by a unit mass emission of gas X relative to the same amount of emission of $CFC - 11$ [54] which is taken as a reference gas labeled F11[39].

$$ODP_X = \frac{\int_0^\infty [\Delta O_3]_X^P dt}{\int_0^\infty [\Delta O_3]_{F11}^P dt} \quad (2.4.1)$$

With $[\Delta O_3]$ being the global mean total ozone change induced by the perturbation. The superscript P being the pulse emission and the subscript refers to the perturbation compound.

The steady state form of **ODP** is usually used instead of the integral form above and takes the form.

$$ODP_X = \frac{m_{F11} \times \Delta\mu_{F11} \times \tau_X \times [\Delta O_3]_X}{m_X \times \Delta\mu_X \times \tau_{F11} \times [\Delta O_3]_{F11}} \quad (2.4.2)$$

With **mass (m)**, $\Delta\mu$ being the mixing ratio of the perturbation, τ is the lifetime and $[\Delta O_3]$ is the steady-state annual and global mean total ozone change induced by the perturbation [39].

The ODP formula for N_2O becomes:

$$ODP_{N_2O} = \frac{m_{F11} \times \Delta\mu_{F11} \times \tau_{N_2O} \times [\Delta O_3]_{N_2O}}{m_{N_2O} \times \Delta\mu_{N_2O} \times \tau_{F11} \times [\Delta O_3]_{F11}} \quad (2.4.3)$$

2.5 Drivers of N_2O soil emissions

In the research done by [11] it has been identified that nitrous oxide emissions increases linearly with the amount of mineral fertilizer applied. They said that doubling the [Nitrogen \(N\)](#) fertilizer rates from 100 to 200 kg N/ha increased N_2O emissions by 60%.

In a field study they did near Quebec City, Canada; they found that increasing the N fertilization rate for corn by 50%, i.e from 120 to 180 kg/ha increased the denitrification rate ($N_2O + N_2$) by 40 to 130% and increased the N_2O emissions rates by 50 to 200%. It was also found that N fertilization is the main factor controlling the emission of N_2O .

[38], in their revised study on *greenhouse gas emissions from soils* outline drivers of N_2O soil emissions. The drivers are **humidity, temperature, Exposure and air pressure, vegetation fires, soil pH values, nutrients and vegetation type.**

2.5.1 Humidity

Humidity is considered as the most important soil parameter for soil gases emission. It controls microbial activity and all related processes. N_2O producing bacteria require anaerobic conditions. N_2O production has the highest yield around 60% [Water Filled Pore Spaces \(WFPS\)](#) and lowest when WFPS is below 30%[13].

An increase in WFPS above 80% still leads to an exponential increase of N_2O emissions[23]. The research says that long periods of drought can significantly reduce soil emissions.

2.5.2 Temperature

Soil temperature is the most important to explain the variation of trace gas emissions from soils. Soil moisture and temperature can explain 86% of N_2O emissions. A rise in soil temperature leads to higher emission and higher soil respiration rates which are indicators of increased microbial metabolism.

2.5.3 Exposure

The study showed that site exposure i.e (elevation, morphological position and plant cover) influences soil temperature and moisture. So Nitrous oxide emissions are higher in depressions (low pressures) than on slopes and ridges due to soil moisture. Lower air pressure supports higher soil emissions due to reduced counter pressure on the soil.

2.5.4 Vegetation fires

Vegetation fires or fires in ecosystem can affect the greenhouse gas balance of soils, depending on temperature and duration of the fire, where burned areas were showing lower CO_2 and N_2O fluxes than non-burned reference areas for around one month after burning.

2.5.5 Soil pH

[38] also showed that soil [potential of Hydrogen Ion \(pH\)](#) values influence the microbial activity where practices such as liming influence soil emissions. N_2O emissions decrease only under acidic soil conditions and nitrification increases with higher pH values, since the equilibrium between NH_3 and NO_3^- shifts to ammonia.

2.5.6 Nutrients

Availability of nutrients is an utmost need to microbial and plant respiratory processes. Thus, natural N and C content in soil, aside from atmospheric deposition, manure or fertilizer applications have important function [38]. Carbon availability influence the N_2O emissions according to [10].

The study of [38] showed that there is a negative correlation between N_2O and C to N (C/N) ratios, with the lowest N_2O emissions at C/N ratios ≥ 30 (for limited disintegration of organic material) and the N_2O emissions being highest at C/N value of 11 (for optimum disintegration and humus build up). This study showed that nutrients in combination with droughts and pH values, N_2O emissions can be substantially omitted at C/N ratio >20 .

In the same study, it has been shown that the application of liquid manure (urea) led to higher N_2O emissions under aerobic soil conditions whereas NH_4^+ fertilizers caused higher N_2O emissions under saturated conditions.

[38] advised that to minimize the N_2O emissions from agricultural lands, fertilizer application rates need to be adapted to plant needs because not all forms of nitrogen can be taken up by plants. Non plant available N amounts lead to increasing N_2O emissions.

2.5.7 Vegetation

The age of vegetation and the type of trees influence soil respiration. Young spruce forests were noticed to higher respiration rates compared to over 10 year-old stands. This study found that on agricultural sites, N_2O emissions from legume-N were significantly lower than N_2O emissions derived from fertilizers-N.

2.6 Some methods used for N_2O assessment

The methods used in the research of [38] are:

2.6.1 Micrometeorological methods

A 3-D ultrasonic anemometer and a gas analyser attached to a tower or mast of at least a 2m height are needed for this method. But also Open Path [Fourier Transform infrared \(FTIR\)](#) Spectroscopy is another near ground micro-meteorological method, using an instrument that can be mounted on a tower or a pole to analyse N_2O .

2.6.2 Laboratory experiments methods

This method makes use of field chamber system to analyse N_2O soil emission in the laboratory or in the lysimeter on field.

2.6.3 Airborne measurements

Airborne measurements work with sample collected from of transects (gases over different types of land use or from near surface environments to higher tropospheric altitudes). Samples collected are stored in flasks and analysed in laboratory using gas chromatography for N_2O .

2.6.4 The soil emission modeling

The method was used to model the daily decomposition, nitrification, ammonia volatilization and N -uptake of plants and plants growth. The [Denitrification and Decomposition \(DNDC\)](#) model was used.

[10] used 3 methods to estimate N_2O emissions. The first method used is the [Intergovernmental Panel on Climate Change \(IPCC\)](#) method, which is the most common bottom-up method used to estimate N_2O emission for national inventories. It is based on soil surface gas flux measurements from numerous global sites. Emissions are assumed to be proportional to soil N inputs from various sources and the method accounts for emission from biomass burning and from N -transformations occurring in manure management systems.

The second method used to estimate N_2O emission is the DAYCENT (daily century) model which is a more sophisticated bottom-up approach and it accounts for the influence of other factors such water, temperature, O_2 and labile C availability as well as plant N demand that influence direct N_2O soil emissions.

The DAYCENT model predictions are based on soil surface flux measurements. The third method is the top-down approach that infers anthropogenic N_2O emissions from changes in atmospheric N_2O concentration and N_2O removal rates.

2.7 Mitigation measures proposed for N_2O emission reduction

Agriculture is still the most anthropogenic source of N_2O and the mitigation measures proposed by [31] for the reduction of N_2O emission involve the consideration of the nitrogen cycle of agricultural systems as a whole and target the increase in nitrogen efficiency of the systems. Two main mitigation measures have been found efficient namely the use of nitrification inhibitors and the control of the denitrification end products. The commonly used nitrification inhibitor is [Dicyandiamide \(DCD\)](#) which could reduce N_2O emissions by up to 80%. DCD was found to be inefficient in acidic soils because of its inactivation by binding to humic compounds. The control of denitrification end products, which are mainly N_2O and N_2 , is done by enhancing the conversion of N_2O to N_2 and reduce N_2O emission. [31] in their study say that maintaining a soil pH at 6.5 might help maintain a low N_2O mole fraction or N_2O - N_2 ratio from denitrification.

The [DCD](#) is not the only used Nitrification inhibitor. The use of nitrification inhibitor [3,4-Dimethylpyrazole phosphate \(DMPP\)](#) lead to decreased N_2O emissions from N fertilization, the inhibitor works by inhibition of oxidation of ammonia (NH_3) to nitrate (NO_3^-)[57].

Chapter 3

Methodology

3.1 Site selection

3.1.1 Geographic description of Rwanda

Rwanda is a country located in Central /Eastern Africa at the coordinates $2^{\circ}S$ and $30^{\circ}E$. It has an area of 26,338 square kilometers (10,169 square miles) and is the 149th largest country of the world. Rwanda is bordered by Democratic Republic of Congo to the west, Uganda to the north, Tanzania to the east and Burundi to the south [2].

Rwanda is dominantly high altitude with the lowest altitude of 950 m above sea level, recorded in the South-West of the country.

Mountains are abundant in the central and western parts of the country and they are part of the Albertine branch of the East African Rift. The altitude in this region ranges from 1500 m to 2500m above sea level, with the highest point being at the top of volcano Karisimbi with an altitude of 4507 m.

The eastern part of Rwanda is around 1400m above sea level and is composed of savanna and plains [14].

Though it is in the tropical belt, Rwanda experiences a temperate high land climate due high elevations. The North-Western, mountainous part of the country is cooler than the eastern low-lying part. Rwanda has a daily average minimum temperature that varies between $10^{\circ}C$ and $16^{\circ}C$, whereas its daily average maximum temperature varies between $20^{\circ}C$ and $28^{\circ}C$.

The rainfall in Rwanda is correlated with altitude. The highest parts of the country get most rainfall. Karisimbi receives the highest annual average rainfall of 2,200 millimeter and north of Nyagatare along the Uganda border receives the lowest rainfall with an annual average rainfall of 850 millimeter. The western half of Rwanda receives nearly double the rainfall of the low lying eastern part of the country [32].

3.1.2 Site Description

The mole fraction data of N_2O used in this research are recorded from Mugogo Climate Observatory, station for the year of 2017 .

Mount Mugogo is located in the Northern province of Rwanda.

The geographical coordinates of the site are 1°35'02" South, and 29°33'54" East and it is located at an altitude of 2590 m above sea level. Mount Mugogo is located in a rural area at about 70 km away from the capital city of Rwanda, Kigali and 13 km away from the nearest town of Musanze.

The low infrastructures costs and accessibility of mount Mugogo also motivated the choice of this site [14].

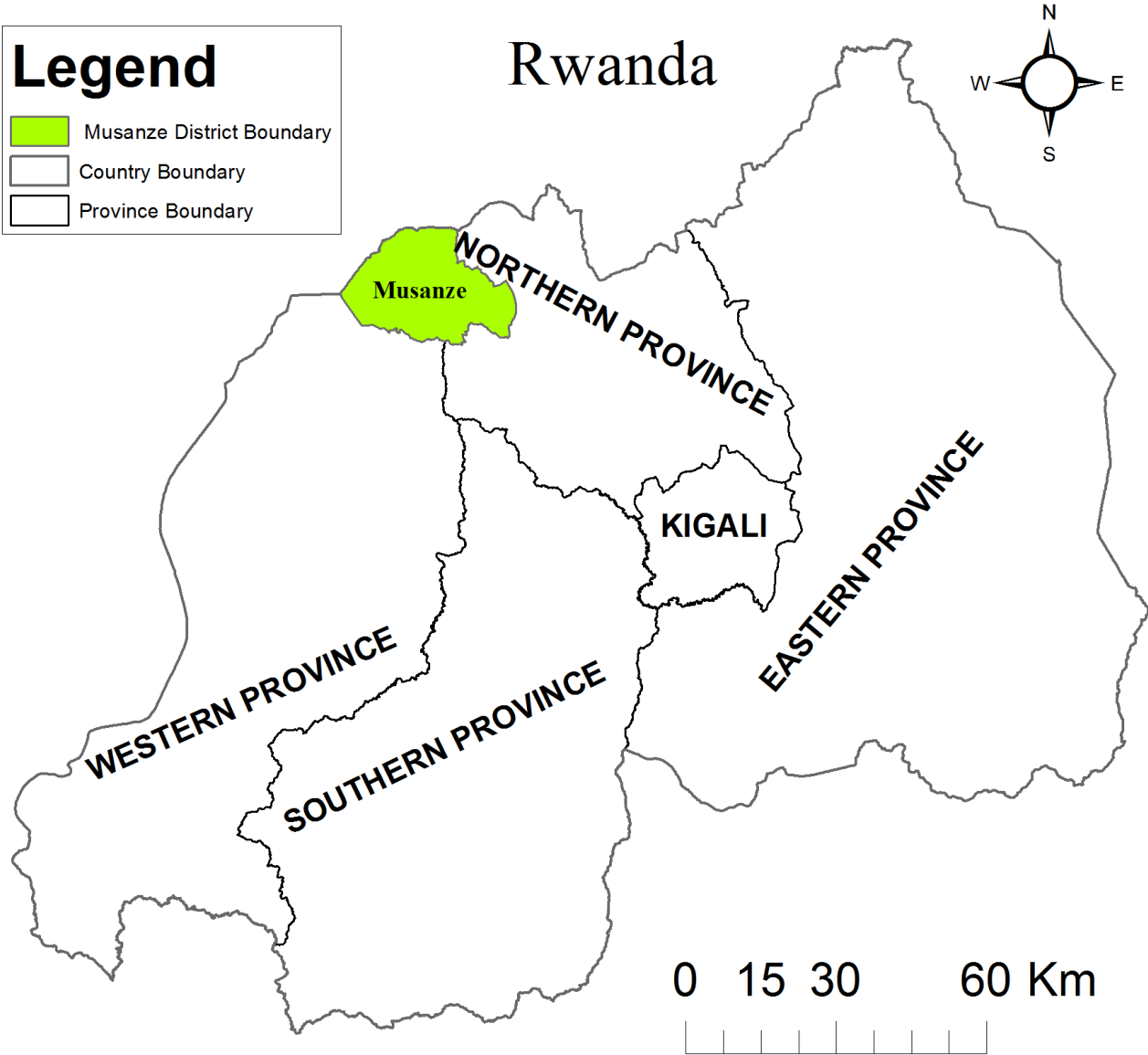


Figure 3.1.1: Map of Rwanda with data site located in Musanze district

3.2 Measurement of Nitrous Oxide concentration

3.2.1 Introduction on Picarro G5105

The mid-infrared analyzer(Picarro G5105) is the instrument used to record the mole fraction data of N_2O . It is a high precision instrument that reliably measures nitrous oxide (N_2O) concentrations and H_2O in the air. It enables stable and precise measurements continuously from the air or from small volume samples.

The mid-IR G5105 system is comprised of two components. The **Pump Power Unit (PPU)** contains a vacuum pump, an 18 V power supply for the laser, and a control module for the ring down detector.

The analyzer also called **Data Acquisition System (DAS)** contains the optics, computer, and electronic cards to power and control the system. In addition, a keyboard, mouse, and video monitor are required to operate the G5105.

The [Figure 3.2.1](#) is the image of Picarro G5105.



Figure 3.2.1: Mid-Infrared analyzer Picarro G5105 [16]

3.2.2 Power requirements

1. Under normal operating conditions, the analyzer consumes electrical power of about 250W. For initial warm up of the system, more power may be required.

2. It is recommended to have power source of 500 w or more for the instrument
3. Make sure the electrical source is clean in terms of “no spikes or outages”.

3.2.3 Chiller Refill Procedure

Chiller with a 40ml water buffer tank is installed for reliable and efficient laser cooling.

- The water in the buffer tank is drained when shipped. Follow the instruction for initial water fill.
- Instrument must be powered off for water tube reconnections and water fill, refill, or drain.
- Every half a year, customers are recommended to inspect the water levels in the buffer tank and add or refresh water in case of water loss or contamination.

Following is a procedure for chiller initial fill:

1. Once the analyzer main box and PPU unit are unpacked and seated level on table.
2. Inspect the water buffer tank for possible crack, and/or external water tube breakage. Fix it before water fill.
3. The black top lid of water buffer is set about 1/2 inch higher than the top of the main box. Adjust the tank level, if necessary.
4. Rotate the black top lid count-clockwise to open the tank, fill the buffer tank with 30ml distilled water, secure the top lid and seal the tank.
5. Disconnect the two hooked up quick connectors, and plug them into the two quick adaptors at the bottom of the buffer tank.
6. Now, you can power on the analyzer to check the water flow. In a normal chiller circulating condition, one water jet should be visibly seen from the 1/8” Teflon tube inside the tank.

Following is the procedure for chiller refill:

1. Power OFF the instrument.
2. As the step 4 of the initial water fill procedure, add water to 80% of the tank capability.
3. Turn on the instrument and circulate water for 5-10 minutes, check the water level and flow.
4. In case water is contaminated, drain the water from buffer tank and replace with fresh distilled water in step 2. Repeat steps 1-4-3, if necessary.

3.2.4 Repeatability

In order to understand the smallest mole fractions or concentration of N_2O that can be resolved by an instrument, repeatability is calculated. Repeatability indicates the precision of the instrument [14].

For N_2O measurements, repeatability is assessed by taking the average standard deviation for each of the three tanks used, namely: CB10203, CB10893 and CC114957.

The average standard deviations found are 0.2343 ppb, 0.3040 ppb and 0.2973 ppb for CB10203, CB10893 and CC114957 tanks respectively. The average for the the three tanks are found to be 0.279 ppb.

Histograms showing the standard deviations for the three tanks are shown in the [Figure 3.2.2](#) below.

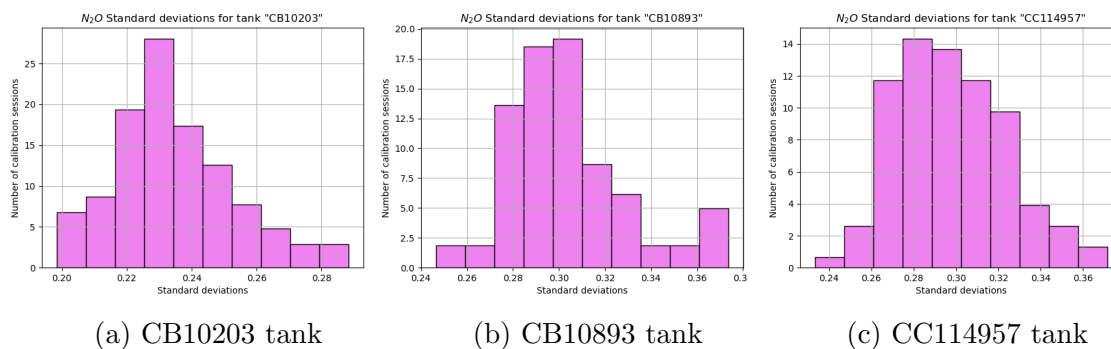


Figure 3.2.2: Histograms of standard deviation during clibration sessions for N_2O for each of the tanks.

3.3 Data analysis

In the presentation of the data, two software packages are used:

Python: this package has helped in the generation of timeseries plots to present the data.

Hysplit Model: [Hybrid Single Particle Lagrangian Integrated Trajectory \(Hysplit\)](#) model:

Is a computer model that is used to compute the trajectory of an air parcel and dispersion or deposition of atmospheric pollutants. It was developed by [National Oceanic and Atmospheric Administration \(NOAA\)](#), USA and Australia's Bureau of Meteorology. One popular use of **HYSPLIT** is to establish whether high levels of air pollution at one location are caused by transport of air contaminants from another location [37].

3.3.1 Statistical Analysis

During the manipulation of the N_2O data it is necessary to make a call to some statistics to be able to get a meaningful idea about the data.

The calculation of the annual mean N_2O concentration is done using the simple formula of statistics for the calculation of mean. The [standard deviation \(STD\)](#) and [correlation coefficient \(R\)](#) are also calculated.

3.3.1.1 Calculation of the mean value

The mean value is a measure of the center value of the data. It is defined to be the sum of the measurements or data values divided by the number of observations or the number of measurements [29].

$$\bar{X} = \frac{1}{n} \sum_{i=1}^n X_i \quad (3.3.1)$$

Where \bar{X} is the mean value, n is the number of N_2O data values considered and X_i is any starting value of N_2O .

The formula enabled me to calculate the seasonal mean values of N_2O concentration.

The seasons considered are [JF](#) season, [MAM](#) season, [JJAS](#) season and [OND](#) season.

3.3.1.2 Calculation of the standard deviation

The [STD](#) is the square root of the variance. It yields a measure of variability and it has the same unit as the original data [29].

$$STD = \sqrt{\frac{\sum_{i=1}^n (X_i - \bar{X})^2}{n - 1}} \quad (3.3.2)$$

With n being the sample size.

3.3.1.3 Calculation of correlation coefficient

The Pearson correlation coefficient is calculated to find that there is a significant relationship between the two variables. In this research the coefficient helps in finding the relationship or any correlation between N_2O concentration and either the [RH](#) and/or the [T](#).

The formula used to calculate the [R](#) is the following:

$$R = \frac{\sum_{i=1}^n (X_i - \bar{X})(Y_i - \bar{Y})}{\sqrt{\sum_{i=1}^n (X_i - \bar{X})^2 \sum_{i=1}^n (Y_i - \bar{Y})^2}} \quad (3.3.3)$$

With positive values of R indicating the increase of X with increase in Y values, whereas negative values of R indicate the increase of X with the decrease of Y values and vice versa. R is nearly zero when either there is no relationship between the changes in X and changes in Y variables or there exists a non linear relationship between the two variables[29].

X variable corresponds to N_2O concentration whereas Y variable can either be RH or T.

Chapter 4

Results and interpretation

4.1 Annual and seasonal mean, and standard deviation

4.1.1 Annual and seasonal mean

After applying the formula for the calculation of mean value, the results of mean annual and seasonal N_2O concentration are summarized in the [Table 4.1.1](#) below:

Table 4.1.1: Annual and seasonal mean values of N_2O concentration for the year 2017

Time Period	Annual	JF	MAM	JJAS	OND
Mean N_2O in ppb	329.5	329.79	329.08	329.30	330.15

4.1.2 Standard deviation

The value of [STD](#) for N_2O concentration is found to be 0.6965 ppb. The value of standard deviation is close to the one found by [27] which is 0.71ppb for detrended N_2O time series.

4.2 Correlation coefficients

The correlation coefficients between N_2O concentration and Relative humidity and temperature are calculated using the formula for [R](#) calculation.

The [Table 4.2.1](#) below shows the values obtained.

Table 4.2.1: Values of Correlation coefficients of N_2O versus RH and N_2O versus T

Parameter	N_2O Vs RH	N_2O Vs T
R value	-0.1325	0.1085

4.3 Interpretation of the results

With the help of python, data are presented in terms of plots. Time series plots are generated and are good tools to understand the seasonal variability for N_2O concentration. Bar plots helps to compare N_2O concentration for the seasons.

The [Figure B.0.2](#) shows that the concentration of N_2O for the year 2017, has an increase pattern except during march, April and May. The same observation was found by [25].

From [Figure B.0.3](#), the concentration of N_2O tends to be stable from January to February. The concentration increases almost as much as it decreases. Therefore there is no much fluctuation in **JF** season's daily N_2O concentration. The increase in concentration goes on from May to December according to the figure. During **JJAS** season, N_2O concentration increased as well as during the **OND** season. [Figure B.0.3](#) also shows that the daily concentration generally started to drop from March up to around May.

From [Figure B.0.4](#), the concentration of N_2O doesn't change much from January to February. The increase in N_2O concentration is almost equal to concentration decrease. The figure also shows that the daily concentration generally started to drop from March up to around May. [Figure B.0.4](#) depicts the general decrease in N_2O concentration during the **MAM** season when compared to the previous season, which is **JF** season. The increase in concentration went on from May to December. During the **JJAS** season, as well as during the **OND** season N_2O concentration increased.

[Figure B.0.5](#) shows that the monthly concentration decreased from February to May and increased during the remaining months of the year. The increase in concentration goes on from May to December .

The N_2O concentration increased during **JJAS** season, as well as during the **OND** season.

The [Figure B.0.6](#) above is the bar plot that shows the comparison in terms of N_2O concentration among the four seasons in Rwanda, namely the short dry season or **JF** season, the long rainy season or the **MAM** season, the long dry season or the **JJAS** season and the short rainy season or **OND** season.

According to the figure, the concentration decreased during the **MAM** season when compared to the previous season, which is **JF** season.

The mean N_2O concentration for the **JF** season, **MAM** season, **JJAS** season, and **OND** season are 329.79, 329.08, 329.30 and 330.15 *ppb* respectively as shown on **Figure B.0.6**. The soil may have been saturated during **MAM** season, with **WFPS** of greater than 60 % which may have implied the denitrification to be the only dominant process in N_2O emission [7]. According to the development of agriculture which requires the use of nitrogen fertilizers, the concentration of N_2O should increase overtime because agriculture contributes to two third of N_2O emission in the atmosphere [9].

Nitrogen addition stimulates N_2O emission during the growing season and even nitrogen addition and rainfall reduction increases the N_2O emission by more than 50 % [46].

During the long dry season, or the **JJAS** season, we expect to experience the least rainfall. The agricultural season C takes place in the period from July to September [36]. It is expected that Nitrogen fertilizers are added to soil and that fact may have made the N_2O concentration to increase from 329.08 *ppb* during the long rainy season **MAM** to 329.30 *ppb* during the long dry season **JJAS**.

The input of crop residues can also stimulate microbial activity and lead to N_2O emissions . The soil organic matter plays a direct and indirect role on denitrification process as most of the denitrifying bacteria are heterotrophic.

The soil temperature positively affects the functioning of microbial communities and therefore affects N_2O emissions from the soil.

During the long dry season in different parts of the country, soil management practices such as fertilization type and quantity, tillage and ploughing usually take place and such practices have an impact on N_2O emission [7].

The **Figure B.0.7** shows that N_2O concentration is positively correlated with weather temperature and negatively correlated with weather humidity.

Figure B.0.8 shows that during the **MAM** season a decrease in concentration was experienced while the previous season, which is the **JF** season, a concentration increase was experienced. The **Figure B.0.8** shows that for both the **JJAS** and the **OND** seasons, N_2O concentration increased.

The soil water content is one of the factors that govern the N_2O emissions. But the soil temperature when coupled with high rainfall events lead to bursts in N_2O emissions .

So the reason for the decrease of N_2O concentration during the **MAM** season is probably the highest rainfall amounts during the year 2017 which is not coupled with soil temperature [8]. During the **MAM** season the weather temperature dropped compared to other seasons according to the bar plots shown in [Figure B.0.9](#) .

The [Figure B.0.10](#) shows the rose plots of wind direction. During the period December-January-February, the sun is over southern part of Africa, which leaves the northern part very dry. The dryness results in biomass burning with some caused by people while preparing their lands for agriculture [14].

The biomass burning in northern, especially north-eastern part of Africa as indicated by the wind rose plots in [Figure B.0.10](#), can have an influence on the observed rise in N_2O concentration during the **JF** and **OND** seasons because N_2O has a long lifetime and can travel quite long distances away from the source.

The hysplit trajectory model accessed from the NOAA website, by using the backward trajectories, shows that most of the trajectories came from south-east of Rwanda. The trajectories that made it to the data site could come as far as from Madagascar as can be seen on [Figure B.0.11](#). The [Figure B.0.11](#) shows that pollution can be globally transported.

4.4 N_2O Correlation with temperature and with humidity

The values of correlation coefficients of N_2O with temperature and with humidity are found to be 0.1085 and -0.1325 respectively. The values indicated a positive correlation between N_2O concentration and temperature, and a negative correlation between N_2O concentration and humidity.

The correlation among the variables is depicted in figure [Figure B.0.7](#).

The low values of the coefficients show that both negative and positive correlation between the variables, namely N_2O concentration and humidity, N_2O concentration and temperature respectively is not significant and is therefore weak. Humidity and N_2O concentration are therefore negatively correlated and the increase of one implies the decrease of the other and vice versa.

The higher correlation was found to be with the temperature. This has a sense since the microbial activity during denitrification increases with the increase of temperature [46].

Chapter 5

Conclusion and Recommendation

5.1 Conclusion

During the assessment of seasonal variability of nitrous oxide, using the data measured with the mid infrared analyzer instrument also known as Picarro 5105, it has been found that the N_2O concentration increased for the majority of the year 2017. The increase started from May up to December, which is added to the increase experienced during a short period of January-February season. It has been found that the concentration increased during the dry season due to regional, continental as well as global biomass burning. The research by [22] found that emissions from fire areas can be transported thousand kilometers and such emissions are not to be taken as local problem only. Since Nitrous oxide is a long lived species, its continental transport is very possible and therefore the concentration of N_2O found is not necessarily produced, sourced in Rwanda. The measured N_2O concentration is the combination of downwind transported local and regional N_2O ; and that from the other parts of the continent. The rose plots showed that the majority of the wind for the year 2017 was blowing from East, specifically north-east and south-east.

5.2 Recommendation and future work

Due to the threats, of global warming, ozone layer depletion and climate change induced by the greenhouse gases, of which N_2O is a part, with negative impacts to the health and safety of beings and materials, further research is needed to understand the seasonal variation of N_2O .

Since data of N_2O for one year were used during this research, there is a need for future works to focus on seasonal variation of N_2O concentration using data of longer periods of time in order to elucidate the effect of rain and dryness to N_2O emission. The estimation of inventories for N_2O emission in Rwanda is of utmost importance for it will allow to know the most contributing source of the greenhouse gas, N_2O , and therefore will help in quantifying the N_2O concentration that comes from outside of Rwanda.

The knowledge of separate sources would help in policy and decision making, and will be a tool to be used when putting in place mitigation measures to reduce the N_2O emissions.

Appendices

Appendix A

Table of Monthly-mean values for T, RH and N_2O

Table A.0.1: Monthly-mean values for T, RH and N_2O

Date	Monthly-mean value		
	Temperature in °C	Humidity in %	N_2O in ppb
1/31/2017	13.62046	79.01096	329.7553
2/28/2017	14.14096	79.27361	329.8184
3/31/2017	-3.12404	76.12093	329.5627
4/30/2017	13.25356	92.98105	328.9459
5/31/2017	12.35471	94.836	328.7018
6/30/2017	13.52935	83.33763	328.9798
7/31/2017	13.90739	73.96556	329.2898
8/31/2017	13.82501	78.99482	329.3403
9/30/2017	14.02463	79.2246	329.5948
10/31/2017	14.23098	79.95216	329.8138
11/30/2017	13.11978	87.63064	330.2314
12/31/2017	14.06337	80.14874	330.6572

Appendix B

Figure plots related to seasonal variability of N_2O , 2016

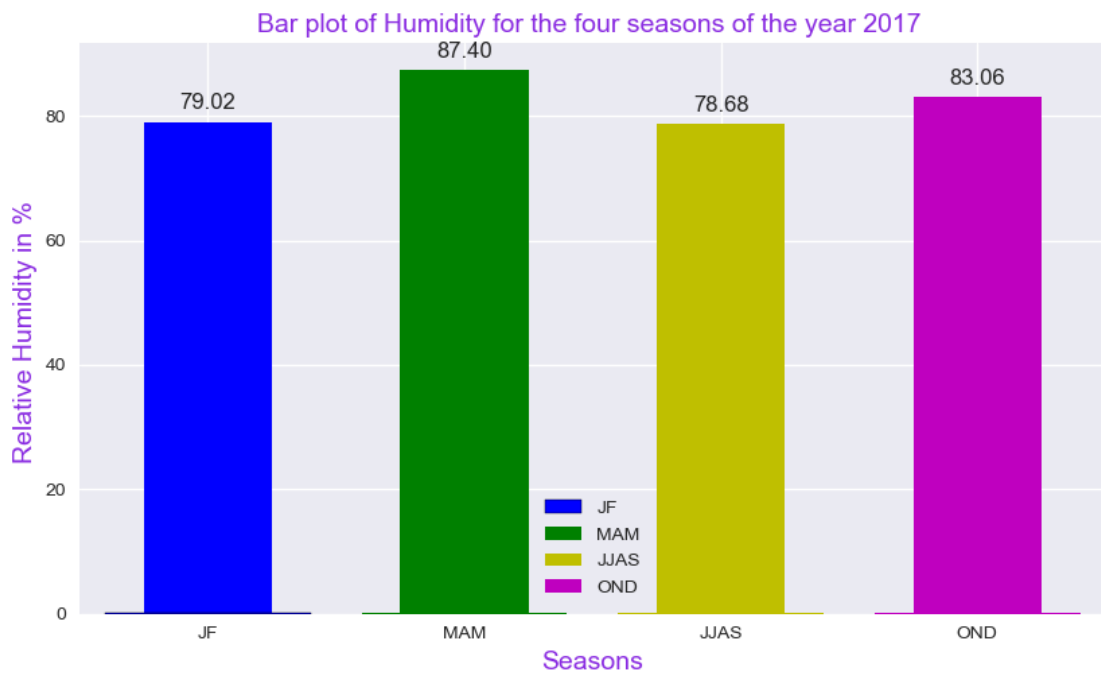


Figure B.0.1: Relative humidity at the data site

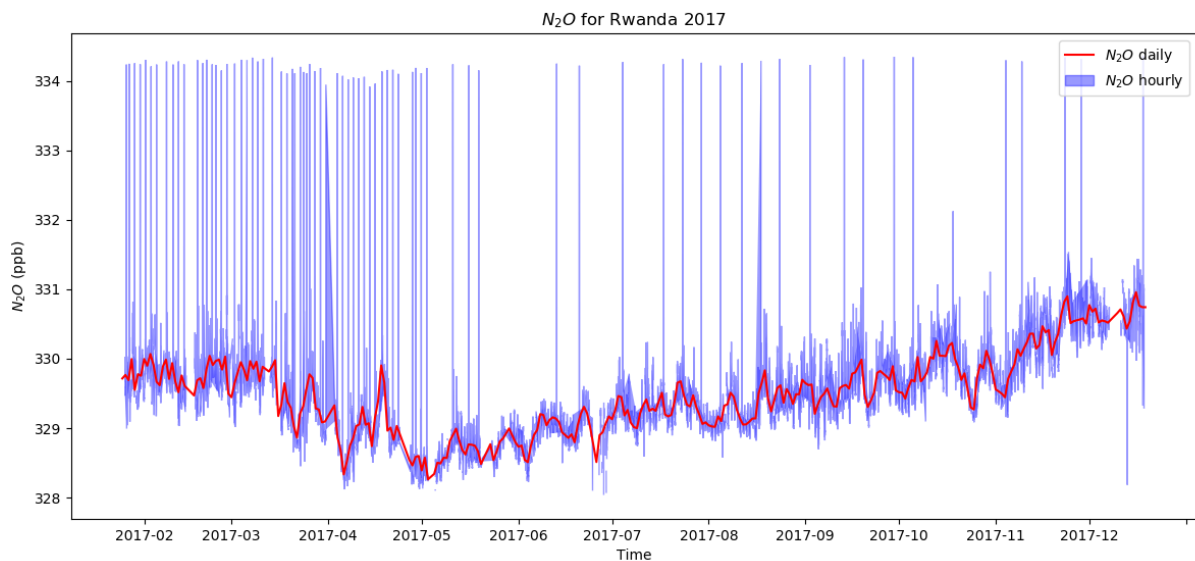


Figure B.0.2: Filled plot of hourly and daily N_2O concentration for 2017

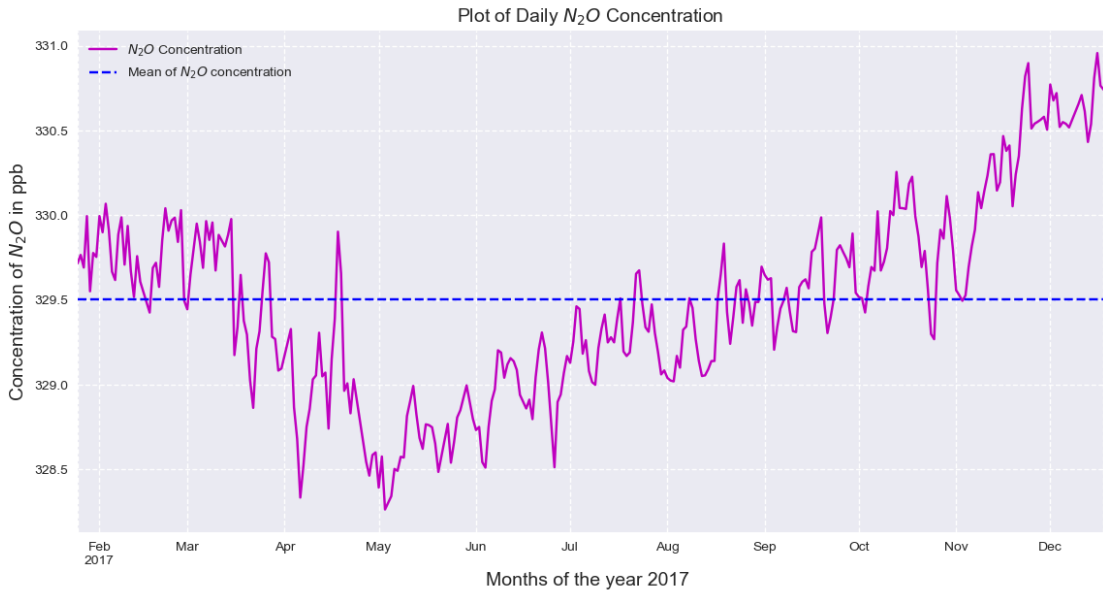


Figure B.0.3: Daily-mean N_2O concentration

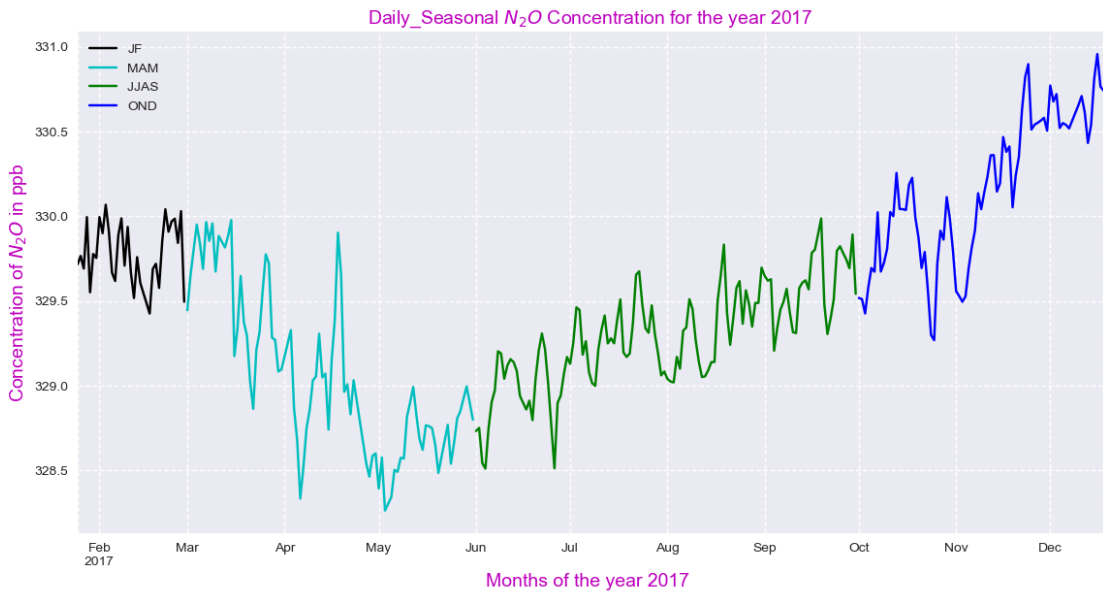


Figure B.0.4: Daily-seasonal N_2O concentration

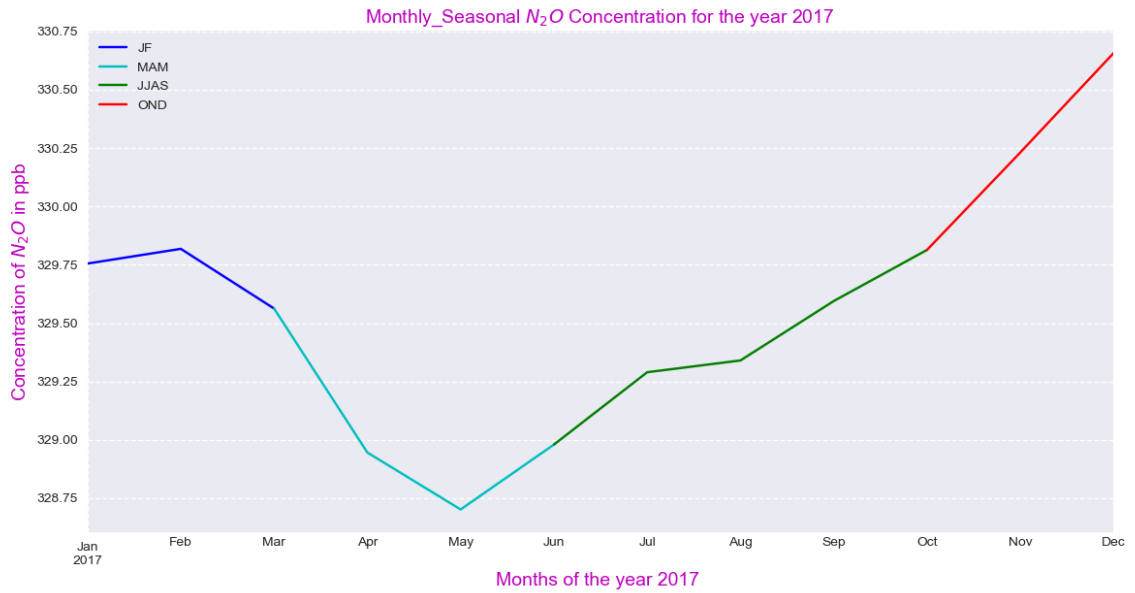


Figure B.0.5: Monthly and seasonal variations of N_2O concentration during the year 2017

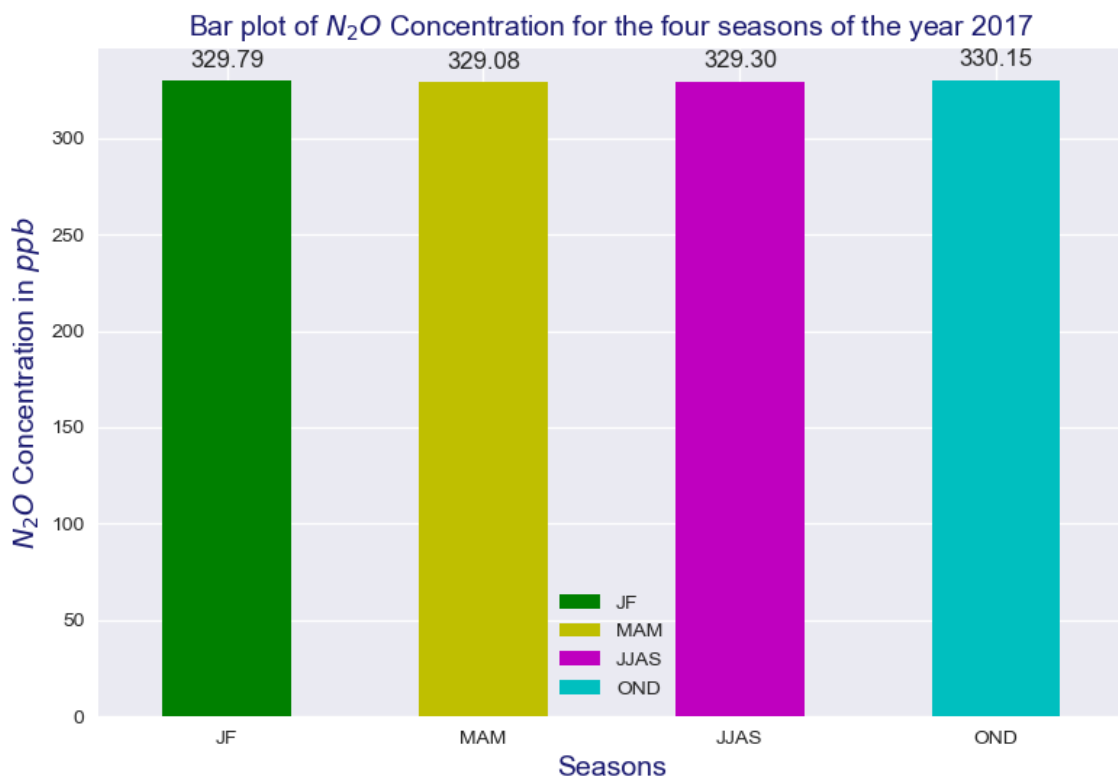


Figure B.0.6: Comparison of N_2O concentration by Seasons

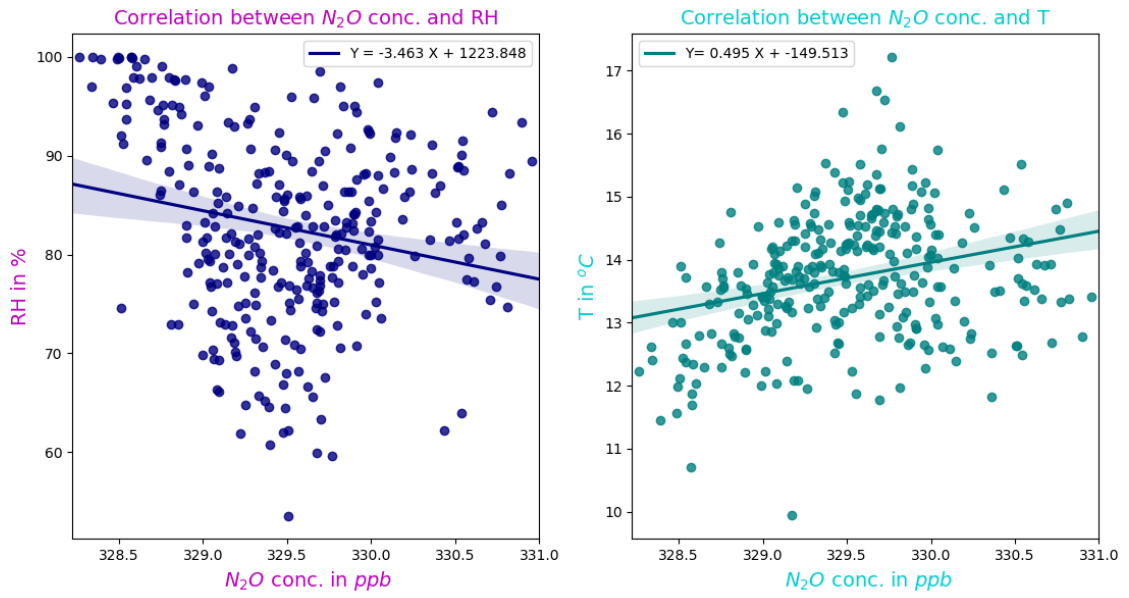


Figure B.0.7: Correlation between Humidity, Temperature and N_2O Concentration



Figure B.0.8: plot of seasonal N_2O concentration variation

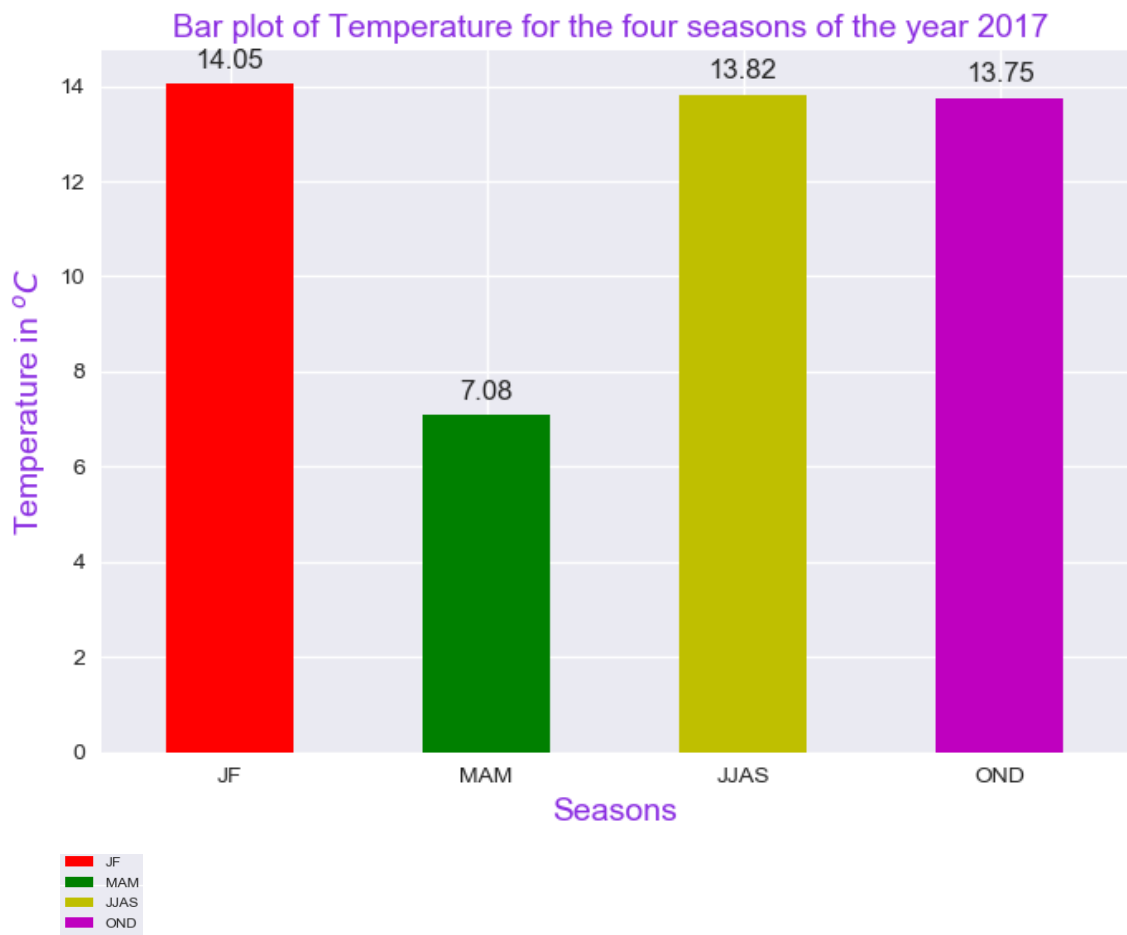


Figure B.0.9: Bar plot for Temperature at the data site

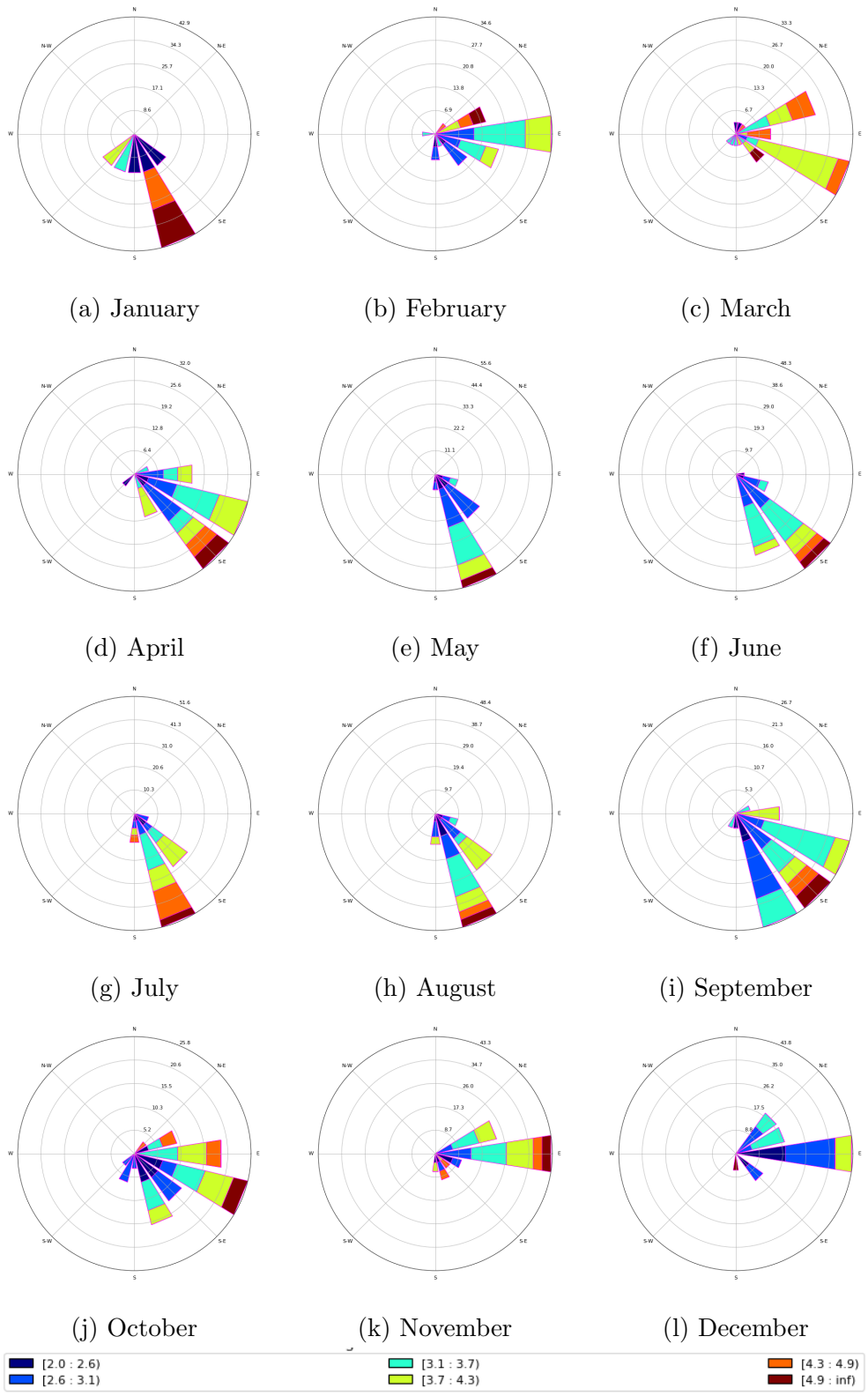


Figure B.0.10: Rose plots of the wind at Mugogo for the year 2017

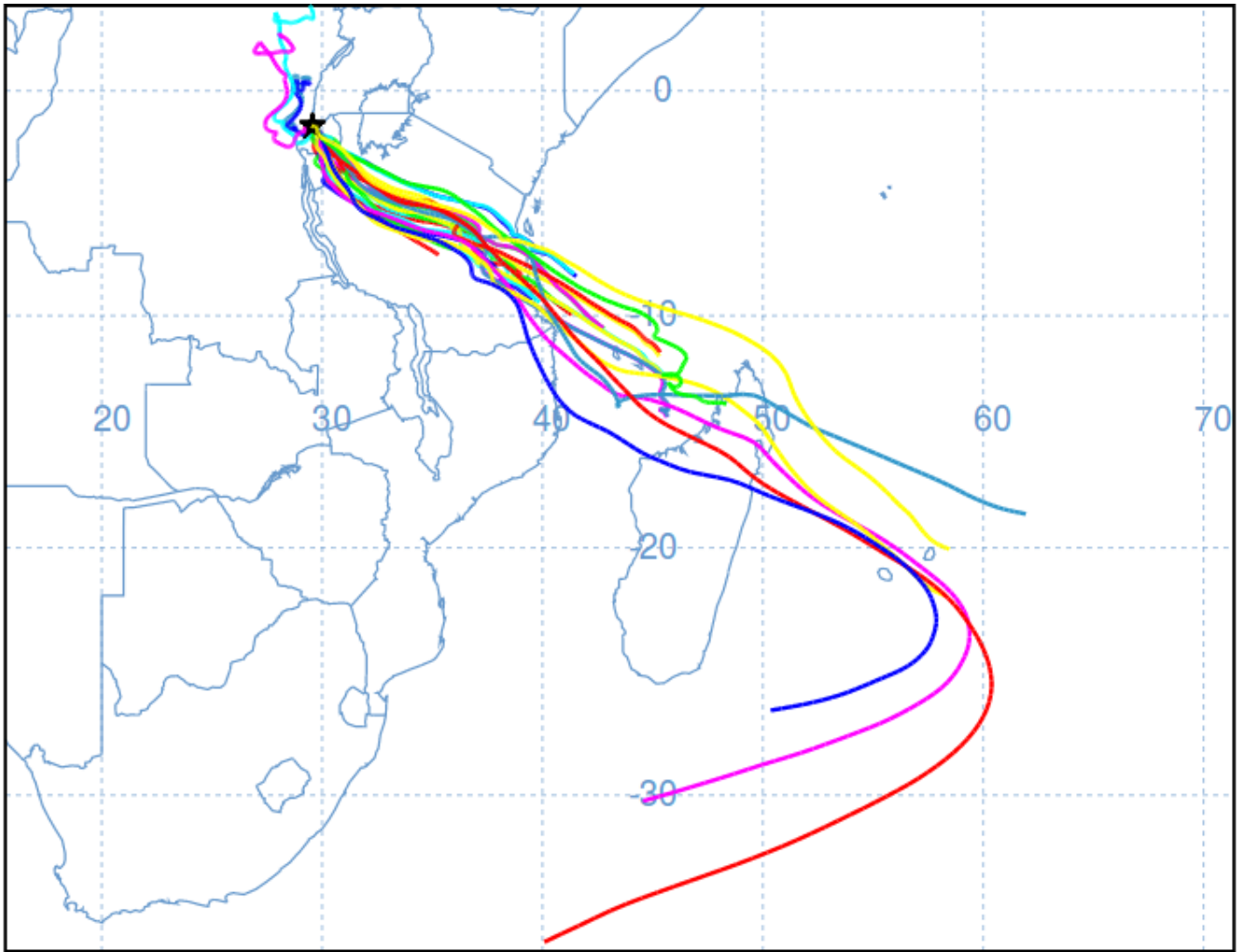


Figure B.0.11: Hysplit Backward trajectories at Mugogo starting from 2018 and from 500 m AGL, generated from NOAA website

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