# Validation of an Improved Soil Depositional Mass Balance Model

# Using Cs-137 as a Tracer of Deposition in Cultivated Soils

**Master Thesis** 

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## Introduction

Nowadays soil erosion is a serious environmental problem. Surface layers of agricultural fields are depleting and reducing the land productivity, while hunger is a major concern in wide areas of the world. The necessity to control and predict erosion led to the development of different estimation methods.

Erosion studies started with traditional methods which were time consuming and expensive. Afterwards use of fallout radionuclides became an efficient alternative for conventional methods.

Cs-137 was produced during the nuclear weapon tests and because of its long half life (30.02 years) and strong fixation by fine soil particles; it became a valuable tracer for erosion studies. Later, on 1986 the Chernobyl accident increased the Cs-137 budget in local scales. The Cs-137 method requires information about the local atmospheric deposition which produces a reference value for an undisturbed site and also a mathematical relationship to estimate the values of erosion or deposition rate quantitatively for each study site. There are many models available for this means and the result of estimations strongly depends on the model which is chosen and the parameters used in that model. This study is devoted to validation of one of these mathematical models which is especially designed for estimations of deposition rates and is called Improved Depositional Model. This model was presented by Jha & Kirchner in 2009 and is an improved form of the simple depositional model (Zhang et al. 1990). In the improved form, the effect of ploughing was added to the estimations of deposition rate. This master thesis investigated the parameters and assumptions involved in this model and estimated its precision. In addition to the validation, in this study a more detailed form of the model that includes more natural processes is introduced. At the end the results of the improved depositional model and its detailed form are compared with each other and experimental data.

#### **1. Soil Erosion**

Throughout the Earth, wherever there is soil, the land is subject to erosion. It means soil and other earth materials are being detached from one area, transported different distances and deposited on another area; Therefore, Earth's surface landscape changes its shape over time.

Soil erosion is occurring naturally and also as a result of human activities on land. The natural agents which are wind and water contribute a significant amount of soil loss each year. On the other hand anthropogenic agents such as deforestation, overgrazing, changes in land use, and non-sustainable farming practices have accelerated the soil loss to the point that today the rate of soil erosion exceeds the rate of soil formation over wide areas.

The formation of soil happens over a very long period of time by weathering of rocks and minerals. Remnants of plants and animals develop the top layer of soil and make it productive. During the erosion processes this layer of soil is depleting. This depletion reduces the fertility of soil and crop production. On the other hand, deposition of surface material in lakes and reservoirs can reduce the quality of surface waters.

Today erosion is becoming a major problem all over the world and considering its important effects on human life, it is necessary to study it, predict it and find the ways to control it. Erosion processes by each agent are explained in the following.

#### **1.1. Erosion by water**

Water is the most important cause of erosion in most of the Earth. Water erosion mainly happens during the rainfall. Water drops hit the surface layer of soil, detach them and disperse them. Obviously the more intense is the rainfall the more particles especially large ones are being washed away by the force of runoff waters.

On the other hand there are some parameters which have significant effect on the extent of erosion event after each rainfall. One example is the steepness of the slope. Naturally, the steeper the slope of a field, the greater the amount of soil loss by water will be. Vegetation cover is another important factor in erosion induced by water. Vegetation cover works as a protection shield for land. It reduces erosion by protecting the soil from intense rainfalls and slows down the runoffs. Naturally, fields are not covered will vegetation completely and also all the year. Therefore, plants which cover the earth better and stay longer reduce the amount of erosion more effectively. Though the tillage practices usually disturb the natural cover of fields and accelerate erosion in most cases.

#### 1.2. Erosion by wind

The speed and direction of wind has a direct relationship with the extent of soil erosion. When the soil is dry, it can be easily transported by wind but in humid climates soil particles cannot be easily removed.

Soil particles size is one of the major factors that control the magnitude of soil erosion by wind. Very fine particles can be detached easily by the wind and transported long distances. Fine and medium size particles can be lifted and deposited, while large particles will be just pushed slightly over the surface.

As it was mentioned for erosion by water, vegetation is like a shield for the land. Dry and bare soil shows the least resistance to wind erosion.

If the erosion happens only naturally, usually an equilibrium condition is developed between soil depletion and production. On the other hand when human activities disturb the environmental conditions, erosion rate may be accelerated significantly. It means top layer of soil is depleting without adequate replacement. As a result productivity of soil decreases sharply and agricultural chemicals are transported and sediment on surface waters and endangers the life of aquatic animals and adversely affects the water quality.

A summary of all the factors effecting the soil erosion is shown in the following picture:



Figure 1. Soil erosion natural and human factors (http://www.nationalarchives.gov.uk/webarchive/)

The first step to manage and avoid these unwanted consequences is to become aware of the erosion rate which means to be able to measure it.

The types of erosion measurements differ for water and wind erosion and depending on the purpose of study there are different techniques to measure each kind of erosion.

The focus of this study is mostly on water and tillage erosion on cultivated lands and use of fallout radionuclides to measure it. The following chapter describes the use of fallout radionuclides as a tracer of erosion and sedimentation.

## 2. Measuring soil erosion using fallout radionuclides

Over the past 50 years, the potential for using natural and man-made radionuclides to study erosion and sedimentation has drawn much attention. Fallout Caesium-137, natural Lead-210 and Beryllium-7 are radionuclides that have been used to perform measurements of erosion rates and patterns independently (Zapata, F., 2002). Each of these radionuclides is introduced in the following.

#### 2.1. Radionuclides

#### 2.1.1. Caesium-137

Radio-caesium (Cs-137) does not occur naturally on the Earth, it is exclusively anthropogenic.

Through atomic bomb testing since 1945, in the 1950s and 1960s radioactive fission products have artificially entered the atmosphere, spread worldwide and gradually deposited on the landscape through wet –precipitation- and dry deposition. Among these fission products Cs-137 was present in high amounts after each bomb testing and due to its long half life; it is a unique global tracer.

The reactor accident in Chernobyl that happened on 1986 also dispersed a great amount of Cs-137 regionally that affect the total global deposition budget as well.

#### 2.1.2. Lead-210

Lead-210 is a natural product of the uranium-238 decay series derived from the decay of gaseous radon-222, the daughter of radium-226. Radium-226 occurs naturally in soils and rocks and will generate lead-210 which will be in equilibrium with its parent. Diffusion of a small quantity of the radon-222 from the soil introduces lead-210 into the atmosphere, and its

subsequent fallout provides an input of this radionuclide to surface soils and sediments which is not in equilibrium with its parent radium-226. This fallout component is called "unsupported" or "excess" lead-210, since it cannot be accounted for (or supported by) decay of the in-situ parent. The amount of unsupported or atmospherically-derived lead-210 in a sediment sample can be calculated by measuring both lead-210 and radium-226 (via lead-214) using gamma-spectrometry and subtracting the supported or in-situ component (Zapata, 2002).

Pb-210 has been constantly deposited on soil over years and this continues fallout makes it a valuable tracer in sediments. On the other hand, due to its high amount of annual deposition and most often small erosion rates, it is not the best tracer for erosion.

#### 2.1.3. Beryllium-7

Beryllium-7 is also a naturally occurring radionuclide produced by the bombardment of the atmosphere by cosmic rays causing spallation of O and N atoms in the troposphere and stratosphere. Production of Be-7 is relatively constant and produces a constant fallout deposition on the landscape as well (Zapata, 2002).

#### 2.2. Radionuclides as tracers of soil erosion

When Caesium-137, Lead-210 and Beryllium-7 reach the soil surface through wet or dry deposition processes, they are quickly and strongly adsorbed by the soil and are almost non-exchangeable in most environments. Therefore wind and water are the dominant agents of moving soil particles containing Cs-137, Pb-210 and Be-7. Basically, these radio-isotopes that are adsorbed to fine soil particles cannot be taken up by plants or react with chemicals, thus they travel with wind and water with the same patterns as soil. It means tracing them is like tracing labelled soil particles (Toy et al., 2001; Zapata, 2002).

Accurately measuring Cs-137, Pb-210 and Be-7 activities in environmental samples is relatively easy. Measuring patterns of the distribution of Cs-137, Pb-210 and Be-7 provide information on average soil redistribution rates and patterns. Based on their half lives, Cs-137 (30.02 years) and Pb-210 (22.3 years) are used to study medium term and Be-7 (53 days) short term soil redistribution rates (Walling and He, 1999).

The Cs-137 technique was chosen to be used for this study. Thus, more information about the applications of this technique in erosion and sedimentation studies will be presented in the following chapters.

#### 2.3. Application of Caesium-137 in soil erosion studies

Caesium-137's high tenacity to become fixed to fine soil particles, its relatively long half-life, and world-wide distribution have made it a universal environmental tracer for studying soil erosion and sedimentation (Ritchie & McHenry, 1990; Walling, 1998; Walling & He, 1999).

The basis of the Caesium-137 technique can be summarised as follows:

Caesium-137 was deposited as fallout primarily during the late 1950s and the 1960s and in most environments was rapidly and strongly absorbed by **fine** soil particles at the ground surfaces.

Since the Cs-137 moves in association with the soil particles, redistribution of the radiocaesium reflects the movement of soil particles. As it was mentioned before, distribution of Cs-137 in soil can be measured using gamma-spectrometry. Cs-137 is measured in terms of activity (Bq) or inventory which is the activity per area (Bq. m<sup>-2</sup>). In any erosion study site, this activity is depleting every year due to radioactive decay and erosion.

To measure the local fallout inventory, it is necessary to find a place in the study field which has not been a subject to erosion or deposition, rather the only reason for degradation of radio-caesium would be the radioactive decay. This study point is called the reference.

The next step will be comparing the reference site's inventory with any other desired points on the same field.

During the nuclear bomb testing, most of the released Cs-137 reached to stratosphere, dispersed worldwide and deposited gradually. Therefore assuming that the initial distribution of the caesium-137 fallout input was uniform is close to reality. As a result, the difference between the measured distribution of caesium-137 from the local fallout inventory and the reference (activity per area) represents the impact of soil redistribution during the period since caesium-137 first deposition.

A higher inventory indicates deposition and a lower inventory indicates erosion.

It means the concentration of Cs-137 in an erosion site which has lost some fine, Cs-137-taged particles is lower than the reference site and it is higher in a deposition site which has gained high concentrated Cs-137-taged soil particles.

If a relationship between caesium-137 loss and gain and soil loss and gain can be established, it will be possible to estimate rates of soil erosion and deposition from Cs-137 measurements. This can be done using conversion models.

The next chapter describes each of the above mentioned steps for a field study in detail and based on the author's own experiences.

## 3. The Cs-137 technique in action

In order to apply the Cs-137 technique to study soil erosion and deposition several processes are involved. In this chapter each process, which has been performed in practice for this thesis, will be described in detail.

#### **3.1. Sample collection and preparation**

#### 3.1.1. Sampling

A quantity cannot be measured for the entire field which is studied; instead it is only possible to sample a small proportion of the study area and then to use statistical methods to characterize the area as a whole.

There are different sampling designs for different research purposes. In sampling design, the number of objects to be sampled and the procedure to select the specific object to be sampled are chosen by the researcher.

After choosing the sampling design, the most important process is to find the sampling points and especially with the Cs-137 technique, as it was mentioned before, finding a reference site is extremely important. The reference site is used to establish the Cs-137 inventory in the study region. The changes in inventory, both at erosion and deposition sites, can be assessed in comparison with the reference.

As mentioned before, the ideal reference site has experienced neither soil loss nor sediment deposition; the inventory reflects only the atmospheric inputs of the Cs-137 and its decay through time. Level sites that do not receive flows from upslope positions are the preferred locations (Zapata, 2002).

The research site which has been chosen for this study is in Augustenfelde; it is located in the north of Berlin and is an agricultural field which has been cultivated every year. This area had been studied before for various research projects (Jha, 2009; Kietzer, 2006). Therefore there were lots of information about the history of the area and possible reference sites and reference inventories.

In this study the focus is on the vertical profile of Cs-137 in the depositional sites. As a result, sampling took place down the slope of the field which is a deposition site. The reference inventory of the study which has been taken place here one year ago has been used (Jha, 2009) and the decay factor was applied to it to find the reference inventory for the sampling date. After identifying the sampling point, soil samples of  $10 \times 15$  cm have been taken from soil surface with thickness of 5 cm up to 50 cm depth and with thickness of 2.5 cm up to 100 cm depth to determine the depth to which Cs-137 may be found in this site and how its concentration changes with depth especially below the plough depth.

Another vertical column with exactly the same layer profile has been taken close to the same deposition point.

In the study field a hole with the depth of 120 cm was dug:



Figure 2. 150 cm deep hole at the sampling point

Layers of 5 cm and 2.5 cm of soil were gathered using scrapper plat and gauge. Each sample was kept in a plastic bag with the information about the profile and depth of sample written on it.



Figure 3. Sampling tools (gauge, scraper plate, meter)



Figure 4. Using gauge and scraper to take samples

All the 60 samples were transferred to laboratory afterwards. A photo of the sampling team of this study at the end of a successful campaign:



Figure5. Sampling team

#### 3.1.2. Sample preparation

The initial stage of sample handling in the laboratory for this study was oven-drying. The weights of all samples were recorded. They were taken out of plastic bags, spread over separated trays and put in an oven (80°C). In the next stage, they had to be grinded. Stones

were taken out of each sample. Again the weights of dried samples without stones were recorded. Caesium-137 is attached mostly to soil particles smaller than 2 mm; in order to measure the concentration of Cs-137 in homogenized samples, they have to pass through a 2mm mesh sieve. In this study the percentage of stones and particles larger than 2 mm was lower than 5%, therefore a grinding machine -set on 2mm size particle- was used instead of a mesh sieve.

The next stage was filling aluminium bottles with dried and homogenized soil samples. All bottles were filled up to 8cm height and their weight was recorded. This part is important because the bulk density will be calculated from these data. The bulk density is central for further calculations of erosion and sedimentation rates.

When all bottles are filled while information about the depth of each sample is written on its bottle, samples are ready to be measured by gamma spectrometry.

# **3.2.** Gamma-Spectrometry measurements and qualitative estimation of erosion or deposition rates (Zapata, 2002)

The radionuclides within the soil emit gamma photons at known energies. In order for a gamma ray to be detected, it must interact with matter; and to be studied it must be recorded. Gamma ray photons interact strongly with the electrons in the atoms of elements which have high atomic number and density. Therefore the detection of gamma rays takes place when it transfers part or all of its energy to an electron and ionizes it. This charged electron collides with other atoms and ionizes them. These free electrons are collected and their energies are measured. The measured current is proportional to the energy deposited in the detecting medium.

Among several methods and various detectors for determining the gamma ray in soil samples, Germanium (HPGe) detectors are the most cost-effective and high-resolution ones. The above mentioned processes are described for this detector in the following:

The gamma-ray of the radionuclides within the soil interacts with the germanium in the detector, which in turn emits signals corresponding to the energies of the incoming photons. The signals from the detector crystal are routed through an amplifier and directed to a Multi Channel Analyser (MCA) system. Here, the signals are displayed as a spectrum in which emission counts are plotted against radionuclide energies. The peak-count information is converted to specific activities (Bq.kg<sup>-1</sup>) using calibration procedures. In the following each process is described in details.

#### 3.2.1. HPGe Detector

These detectors are fabricated from high-purity Germanium crystals. This crystal generates free electrons in response to absorbing energy of gamma rays from Cs-137 in a soil sample. The magnitude of the charge in the crystal is directly related to the energy of the incident gamma ray. The crystal is a semiconductor.

In addition to the electrons that are excited within the crystal as a function of gamma-ray interactions, further excitation can occur as a result of thermal energy. This additional excitation and electron activity produces background noise in the system.

In order to reduce the effect of this phenomenon, detectors are usually operated at very low temperatures. This is achieved by cooling detectors with liquid nitrogen.

There also exists an external background noise. This is generally due to natural radiation in the detector's environment, and usually affects the whole spectral background. A lead shielding around the detector can help to reduce the effects of this external noise.

#### 3.2.2. Preamplifier

The preamplifier takes the produced charge from the detector and integrates and amplifies it to produce a step-function pulse.

#### 3.2.3. Amplifier

The amplifier takes the pulse signal from the preamplifier and considerably magnifies it. It also filters and shapes the incoming pulse to increase the signal-to-noise ratio. This improves the resolution and shortens the response time to prevent overlap between pulses.

#### 3.2.4. High-voltage supply unit

Charge collection within the germanium detector occurs best when a bias voltage is applied across the detector volume. This is usually in the range  $\pm$  1000 to  $\pm$  5000 V and is applied by a high-voltage supply unit.

#### 3.2.5. Analogue to digital converter (ADC)

The analogue signal produced by the detector and shaped by the amplifier needs to be converted to a digital signal. An ADC converts the analogue signal from the amplifier to a digital value.

#### 3.2.6. Multi-channel analyzer (MCA)

The pulses that emerge from the ADC are registered in one of the channels of the MCA. Today MCA is often integrated to the computer system. The MCA performs a number of tasks including collecting and sorting the input pulses, storing those data in a spectrum, providing a format to display the data on the computer screen, and performing some analysis of the data.

#### 3.2.7. Accuracy of Gamma-spectrometry analysis

A typical Cs-137 erosion study involves analysis of soils and sediments activities. The main objective is to achieve the estimation of activity within an uncertainty of  $\pm$  5-10%. Factors like efficiency of the detector, mass of the sample, geometry used for measuring the sample, internal and external background noise and the quality and thickness of lead shielding contribute to the overall level of accuracy obtained.

The samples taken for this study, have been measured in the laboratory of BfS (Bundesamt für Strahlenschutz), using HPGe detectors.

Laboratory analyses of samples using gamma-spectrometry provide the specific activity of Cs-137 (Bq kg<sup>-1</sup>). These data can be used to provide a general indication of the relative rates of erosion or deposition involved and the associated patterns, but the results can only be qualitative.

#### **3.3.** Quantitative estimates of the erosion or deposition rates

Qualitative data will be only of limited value. In order to study erosion and deposition processes, to be able to predict and establish methods to prevent it, there will be a need to derive quantitative estimates of the rates of erosion or deposition. For this reason, conversion models are required. Such a model can be used to convert measured Cs-137 inventories to estimates of the actual rate of erosion or deposition related to the sampling point. Different conversion models for cultivated soil, which is the main interest in this study, are explained in the following chapter.

#### 4. Conversion models – Deposition model

Quantitative assessment of soil erosion and deposition needs a mathematical model of the erosion process. There are many methods existing that may be used to convert Cs-137 inventory measurement results into numerical estimates of soil erosion and deposition (Walling & He, 1999; Quine & Walling & Govers, 1996). These mathematical models can be divided into two main categories: empirical relationships and theoretical models. Using mathematical models requires additional parameters whose values should be chosen carefully; otherwise it can severely bias the results of soil erosion calculations (Poreba, 2006).

In the following some of these models used for cultivated soil will be introduced. Then, we will focus on depositional models and especially on an improved depositional model which is going to be validated based on the sampling results of this study.

#### 4.1. Proportional model

This model is based on the assumption that all deposited Cs-137 is completely mixed within the plough layer and thus the erosional soil loss is directly proportional to the reduction of caesium inventory in the soil profile. The model was developed by Walling and Quine (1990) and the equation can be written as follows:

$$R = 10.\frac{B.d.X}{100.P.T}$$
(1)  $X = \frac{A_{ref} - A}{A_{ref}}.100$ (2)

Where,

R: mean soil erosion rate (kg  $m^{-2} yr^{-1}$ )

- B: bulk density of soil (kg  $m^{-3}$ )
- D: depth of plough layer (m)
- X: percentage reduction of Cs-137 inventory
- A<sub>ref</sub>: local Cs-137 reference inventory (Bq m<sup>-2</sup>)

A: Cs-137 inventory at the sampling point (Bq m<sup>-2</sup>)

T: time elapsed since beginning of Cs-137 accumulation (yr)

P: particle size correction factor – the ratio of the Cs-137 concentration of mobilized sediment to that of the original soil (He and Walling, 1996)

$$P = \left(\frac{S_{ms}}{S_{sl}}\right)^{\nu} \tag{3}$$

Where,

 $S_{ms}$ : Specific surface area of mobilized sediment (m<sup>2</sup> g<sup>-1</sup>)

 $S_{sl}$ : Specific surface area of original soil (m<sup>2</sup> g<sup>-1</sup>)

v: a constant with a value of 0.65

In addition to caesium inventory data, the proportional model requires the bulk density of the soil, the depth of ploughing and particle size correction factor.

The proportional model does not take into account the fact that the presence of erosion leads to dilution of the caesium concentration in the cultivated soil in subsequent years of ploughing. This fact limits the use of the proportional model to very low erosion rates.

#### 4.2. Mass-Balance model

To overcome the limitations of the proportional model, the mass-balance model was proposed (Kachanowski and de Yong, 1984; Walling and Quine, 1990; Walling and He, 1999). The mass balance model is described by the following equation:

$$\frac{dA(t)}{dt} = I(t) - (\lambda + \frac{R}{d.B}).A(t)$$
(4)

 $\lambda$ : the radioactive decay constant for Cs-137 (yr<sup>-1</sup>)

I(t): annual Cs-137 deposition flux (Bq  $m^{-2} yr^{-2}$ )

The model takes into account the actual atmospheric deposition of Cs-137 as well as the dilution of Cs-137 by tillage. Not considering the particle size correction factor and removal of freshly deposited caesium for soil surface before it is mixed into the plough layer are two factors that potentially lead to incorrect estimations of this model (Poreba, 2008).

To overcome problems related to selective sorption and erosion, and removal of freshly deposited Cs-137, the mass balance model was improved by Walling and He (1999) to the form:

$$\frac{dA(t)}{dt} = (1 - \Gamma).I(t) - (\lambda + P\frac{R}{dB}).A(t)$$
(5)

Where,

Γ: fraction of freshly deposited Cs-137 removed before mixing into the plough layer

P: particle size correction factor

With the assumption of total Cs-137 fallout occurring in 1963, rather than its gradual deposition in the period from 1954 to 1976, a simplified mass balance model was derived by Zhang et al, 1990. The mean annual soil loss rate using this model is expressed as follows:

$$Y = \frac{dB}{P} \left[ 1 - \left(1 - \frac{X}{100}\right)^{\frac{1}{(t-1963)}} \right]$$
(6)

Where,

- Y: mean annual soil loss (kg  $m^{-2}$ )
- t: year in which parameter the Cs-137 inventory was measured

#### **4.3. Depositional model**

The basic idea of the depositional model is the same as of the mass balance model. It is particularly used for depositional sites which is our main interest in this study as well.

A simple depositional model which was presented by Zhang et al (1990) describes the concentration of Cs-137 deposited each year by the following equation:

$$C_{d}(t_{l}) = \frac{\sum_{i=1}^{k} C_{e_{i}}(t_{l}) \Delta Z_{i} S_{i}}{\sum_{i=1}^{k} R_{ero_{i}} S_{i}}, \dots l = 1, 2, \dots, n$$
(7)

Where,

K: number of eroding areas

n: year of sampling starting from 1963 (at 1963, n=0)

 $C_{ei}(t_l)$ : Cs-137 concentration mobilized from eroded area  $S_i$  (Bq m<sup>-3</sup>)

 $\Delta Z_i$ : height of soil eroded each year (m)

 $S_i$ : area of the slope for each eroding site (m<sup>2</sup>)

 $R_{eroi}$ : erosion rate for each eroding site (m yr<sup>-1</sup>)

Based on the simplified mass balance model and considering radioactive decay, the Cs-137 concentration carried away from each eroded sites to depositional sites is as follows:

$$C_{e}(t_{l}) = \frac{P}{d} A_{ref}(t_{n}) e^{\lambda(t_{n}-t_{l})} (1-P) \frac{\Delta Z}{d} e^{(t_{l}-1963)}$$
(8)

This formula back calculates the deposition from the sampling date  $(t_n)$  to any desired year  $(t_l)$  between the sampling date and 1963. Thus, the first part gives the reference inventory on  $t_l$  and second part gives information about the amount of eroded soil from 1963 up to  $t_l$ .

At the end, the deposition rate in terms of tons per hectare per year (t  $ha^{-1} yr^{-1}$ ) can be described by the following formula:

$$Y_{dep} = 10.B \left( \frac{A(t_n) - A_{ref}(t_n)}{\sum_{l=1}^{n} C_d(t_l)} \right)$$
(9)

Some important factors are still missing in this model; thus, the estimations may be biased. The effect of ploughing is one of the important neglected factors.

In order to overcome the limitations of the simple depositional model, an **improved depositional model** has been developed by Jha, Kirchner in 2008. This model is the main focus of this study and the aim of this master thesis is to validate this model with experimental data and to estimate its precision.

The improved depositional model is taking into consideration the effect of ploughing on the depositional site.

This model like the simplified mass balance and simple depositional models assumes that the total input of atmospheric Cs-137 has occurred on 1963. It also does not project the Chernobyl accident effect on Cs-137 input like all the other models.

Starting the estimations in 1963, the initial concentration of Cs-137 can be assumed as  $C_0$ . Ploughing will mix the Cs-137 within the plough layer uniformly.

Continuing the calculations for year 1964, a layer of soil with thickness of  $\Delta Z$  will be deposited on the depositional site. The concentration of Cs-137 in this layer is C<sub>e</sub>. During this year's ploughing, this new layer of soil will be mixed within the plough player. At the same time, one layer of bottom soil with Cs-137 concentration of C<sub>0</sub> will not be mixed within the plough layer anymore. Thus, the concentration of Cs-137 in the plough layer for this year can be described by the following equation:

$$C_d(t_0 + \Delta t) = \frac{(C_0.(d - \Delta Z) + C_e(t_0 + \Delta t).\Delta Z).e^{-\lambda\Delta t}}{d}$$
(10)

Where,

d: Plough depth (m)

 $C_d$ : Cs-137 concentration within plough layer in 1964 (Bq m<sup>-3</sup>)

 $C_0$ : initial Cs-137 concentration in 1963 (Bq m<sup>-3</sup>)

 $\Delta Z$ : deposited layer thickness (m)

 $C_e(t_0+\Delta t)$ : Cs-137 concentration within deposited layer of soil on 1964 (Bq m<sup>-3</sup>)

t<sub>0</sub>: starting year which is set to 1963 here

$$\Delta t$$
: 1 year

One year later the Cs-137 concentration in the plough layer will be:

$$C_{d}(t_{0} + 2\Delta t) = \frac{\left(\left(C_{0} + (C_{1} - C_{0}).\Delta Z\right).(d - \Delta Z) + C_{e}(t_{0} + 2\Delta t).\Delta Z.\right).e^{-\lambda 2\Delta t}}{d}$$
(11)

Continuing calculations, the general equation of Cs-137 concentration in the plough layer is as follows:

$$C_d(t_0 + N\Delta t) = \frac{\left(C_d(t_0 + (N-1)\Delta t).(d - \Delta Z) + C_e(N\Delta t).\Delta Z\right)e^{-\lambda.N\Delta t}}{d}$$
(12)

Where,

 $C_d(t_0+(N-1)\Delta t)$ : Cs-137 concentration one year before the sampling  $C_e(N\Delta t)$ : Cs-concentration within deposited layer of sampling year

The complete equation of the improved depositional model is the same as equation (9), but  $C_d(t_1)$  is replaced by equation (11).

$$Y_{dep} = 10.B \left( \frac{A(t_n) - A_{ref}(t_n)}{\sum_{l=1}^{n} C_d(t_l)} \right)$$
(13)

n: year of sampling from 1963 (1963 is set to 0)

#### 4.4. Assumptions

In this study I first have investigated the effects of two major assumptions of all mathematical models that have been mentioned here; First, to assume that all the Cs-137 atmospheric deposition has occurred at once on 1963 instead of its real depositional profile which was during the years 1958 to 1983; Second, not to take into account the effects of the Chernobyl accident (1986) on the Cs-137 inventory.

In order to quantify the error that these two assumptions impose to estimations of erosion and deposition rates, some simulations were done. The simplified mass balance model was the first model chosen to be simulated for this means.

 $1^{st}$  assumption: To estimate the relative error of the first assumption, a profile for gradual atmospheric fallout was assumed. Temporal distribution of Cs-137, used in this study (Kirchner, 1992) is shown in the following table:

Year	Cs-137 (%) out of total atmospheric deposition (100%)
1957 – 1958	9.5
1959	9.5
1960 – 1962	13.84
1963	25.86
1964	11.89
1965	10.82
1966 – 1968	7.13
1969 – 1973	6.24
1974 – 1978	4.08
1979 – 1983	1.13

Table1. Cs-137 deposition from 1957 to 1983 relative to the total atmospheric deposition

A typical erosion rate (0.5 cm/yr), particle size correction factor (1.3) and plough depth (30 cm) were assumed and total atmospheric deposition ( $A_{ref}$ ) was set to 1000 Bq/m<sup>2</sup>. Using the simplified mass balance model, considering decay and inserting the above mentioned constants to the following equation,

$$A = A_{ref} e^{-\lambda \Delta t_i} \left(1 - P \cdot \frac{R}{d}\right)^{\Delta t_i}$$
(14)

The residual of Cs-137 in 2010 was calculated.

All the terms used in equation (14) are the same as before.

In order to find how different the erosion rate would be assuming all the atmospheric deposition had happened on 1963, the erosion rate was back calculated. The simplified mass balance model was used assuming that all Cs-137 had been deposited in 1963. Then, the erosion rate for this case was calculated. Result of this comparison was a 5% relative error of the model which assumes all the deposition on 1963.

Increasing the erosion rate from 0.5 cm/yr to 0.9 cm/yr does not change the relative error significantly. Reducing it to 0.1 cm/yr gives the maximum relative error of 10%.

It can be concluded that using the simplified mass balance model, the 1<sup>st</sup> assumption is a good approximation of the real case.

 $2^{nd}$  assumption: similar simulations were done for the second assumption once without the Chernobyl accident and another time with considering it. As it was mentioned before, the 1<sup>st</sup> assumption does not change the results significantly. Therefore, it was used here for simplicity. The relative activity of Chernobyl Cs-137 to bomb testing Cs-137 was varied between 10% and 100%, a typical erosion rate of 0.5 cm/yr and an initial deposition of A<sub>ref</sub>=10000 Bq.m<sup>2</sup> in 1963 were chosen for the calculations.

From 1963 to 1986 (before the accident), calculations are the same for both cases. When calculating the Cs-137 residual from 1986 to 2010, the difference between ignoring and including Chernobyl accident became clear. The temporal variability of Cs-137 for both cases of ignoring and including Chernobyl accident assuming a relative activity of 50% for Chernobyl (Chernobyl Cs-137=50% of Bomb testing Cs-137), is shown in the following:



Figure6. Cs-137 inventory vs. Time (A) including , (B) Ignoring , Chernobyl accident

For the above mentioned case, including the Chernobyl accident, the residual of Cs-137 inventory in 2010 is 3 times higher than in the simple case without considering Chernobyl. Reducing the relative Chernobyl Cs-137 to 10% and increasing it to 100% increases the

residual of Cs-137 inventory 2 to 5 times compared to the simple case. This shows a considerable difference in results and Chernobyl accident's Cs-137 inventory should be included in estimations of erosion rate, especially in areas highly affected by the accident.

In order to show the results in terms of erosion rate, the back calculating technique was used again. A common erosion rate of 0.5 cm/yr was assumed. The Cs-137 inventory using the model that included the Chernobyl accident was calculated. The results were inserted to the simplified mass balance model that does not consider the Chernobyl accident. Then, erosion rates were back calculated for different relative Chernobyl Cs-137 inventories. Erosion rate vs. relative Chernobyl input is plotted in the following; the erosion rate for the case of Chernobyl Cs-137=0 was set to 0.5 cm/yr as before.



Figure7. Erosion rate vs. Chernobyl Cs-137 input relative to bomb testing input

This graph shows clearly that including Chernobyl Cs-137 input in the calculations make a large difference in the results of erosion rate. For the case of relative Chernobyl Cs-137=50% the erosion rate is equal to 0.3, which means 40% relative error of the model which does not consider Chernobyl. Changing the Chernobyl relative input from 10% to 100%, increase the relative error from 10% to 50%. The erosion rate is being over estimated between 10 to 50 percent by the model which does not include Chernobyl, depending on the amount of Chernobyl accident contamination of the study site.

It can be concluded that including Chernobyl the Cs-137 inventory is essential for erosion rate calculations using the simplified mass balance model. Otherwise, the erosion rate is over estimated significantly.

In order to get closer to our goal, which is validating the improved depositional model, all the above mentioned processes were applied to the improved depositional model as well.

First of all, a profile for the Cs-137 inventory of each year was developed. It means starting from 1957 each year has an atmospheric input (it may be zero for some years), for the next year, the inventory will be the last years inventory which has been decayed for one year plus the atmospheric input of that year. This process is repeated up to present and the result will be a temporal inventory profile. This profile is used as the 'Reference Inventory' in erosion and deposition equations.

For the simulations using improved depositional model, a single eroding site is assumed for simplicity.

 $1^{st}$  assumption: First the effect of gradual atmospheric deposition of weapons fallout was quantified. Using the above mentioned reference profile and table1, concentration of Cs-137 at present was calculated and compared with the original model which considers a single atmospheric input on 1963. A plot of Cs-137 concentration vs. time is shown in the following.



Figure8. Cs-137 concentration vs. time (A) the atmospheric deposition at 1963 (B) Gradual atmospheric deposition

As it is clearly shown in the plots considering the gradual atmospheric deposition will give lower Cs-137 concentration today and the difference between two models becomes larger as the erosion rate increases. Depth distribution of Cs-137 concentration for each of the deposition rates assumed is shown in the following figure.



Figure9. Cs-137 concentration vs. depth for different erosion rates considering gradual weapons fallout

The dependency of depth distribution (location of the peak) to the erosion rate is clearly shown in figure 10. The steeper the line between the peak and current concentrations, the higher the erosion rate will be.

 $2^{nd}$  assumption: Coming to the second assumption, the effect of Chernobyl on concentration profile was studied. In this case, the Chernobyl input was taken as 50% of the weapons fallout, like the simulations for simplified mass balance model.

As above concentrations through time were calculated and residual concentrations for today were compared. Time evolution of Cs-137 concentration for both cases of ignoring and considering the Chernobyl Cs-137 atmospheric input is shown in the following plot:



Figure 10. Cs-137 concentration vs. time (A) without considering Chernobyl (B) with Chernobyl

This time, the difference between both cases is the opposite: considering the Chernobyl gives higher concentrations for today. And the difference between two models decreases with increasing erosion rate.

On the other hand the difference between these two models is due to the effects of plowing and dilution which are considered in the improved depositional model at the same time while the simplified mass balance model only takes into account the dilution effect. Plowing moderates the sudden changes in Cs-137 inventory slightly –like the Chernobyl accident that suddenly increased the Cs-137 inventory- and leads to a smaller difference between the original model (equation (12)) and the above mentioned simulations.

Depth distribution of Cs-137 concentration for each of the deposition rates are shown in the following figures.



Figure 11. Cs-137 concentration vs. depth for different erosion rates considering Chernobyl

Now as a third step, both assumptions will be removed from the model therefore this time both gradual weapons fallout and Chernobyl input are included in the calculations of Cs-137 concentrations. Removal of first assumptions (using gradual fall instead of 1963 fallout) decreased the concentrations and removal of second assumption (Including Chernobyl input) resulted into larger concentrations for today. As a result the removals of both assumptions to some extend cancel out each other. The plot of Cs-137 concentrations vs. time for the case of considering gradual weapons fallout and Chernobyl at the same time is presented in the following and the reduction of the difference between two models in compare with the simulations of only gradual fallout and only Chernobyl is clearly shown:



Figure 12. Cs-137 concentration vs. time (A) original improved depositional model (B) improved depositional model considering gradual weapons fallout and Chernobyl

The depth distribution of Cs-137 for each of the deposition rates is shown in the following:



Figure 13. Cs-137 concentration vs. depth for different deposition rates considering both gradual weapons fallout and Chernobyl

Following the simulations of figure13, we would expect in soil to find a sharp peak in the middle of the vertical concentration profile which is exposing the Chernobyl atmospheric Cs-137 input on 1986 and a smaller wider peak on the bottom of the profile with its maximum on 1963. Depending on the erosion rate and Chernobyl share, the sharpness and depth of peaks are changing.

Our results show that for an average deposition rate of 1 cm/yr the difference between the two models are less than 20%. As the deposition rate increases this difference becomes close to 35% for 2 cm/yr and as it gets lower the difference shrinks to 5% for 0.5 cm/yr.

#### 5. Measurement results

As it was explained in chapter 3, in this study the focus is on the vertical profile of Cs-137 concentrations in the depositional site and samples were taken from a deposition point, up to 100 cm with a thickness of 5 cm from the surface to 50 cm depth and with a thickness of 2.5 cm from 50 to 100 cm depth. The smaller scale for the sampling of 50 to 100 cm depth was chosen to reach higher resolution of Cs-137 concentrations below the plough depth.

Before going into details of the measurement results of this study, a brief look at other studies on the same depositional site will be helpful. Two other studies were performed on this area, in 2004 by Barbara Kietzer (Kietzer, 2006) and in 2009 by Abhinad Jha (Jha, 2009). In the following each of these two studies and the results of their measurements are briefly documented.

#### **5.1.** Prior studies

#### 5.1.1. Kietzer (2004), Jha (2009)

These studies were performed on the same field in Augustenfelde. 10 points were chosen (originally by B. Kietzer, in 2004) to be sampled through the slope with 9 eroding points and the  $10^{th}$  point at the end of the slope as the depositional site. Three reference sites were also sampled.

The 10 sampling points and the reference sites were sampled with approximately the same coordinates in both studies which give the possibility for comparison means as well.



Topography of the sampling slope is shown in the following figures.

Figure 14. Relative height and position of the 10 sampling profiles

First of all take a closer look at the results of the reference sites measurements.

In 2004 study the whole inventory for reference 1 was 4547  $Bq/m^2$ , for reference 2 was 6553  $Bq/m^2$  and for reference 3 was 5152  $Bq/m^2$ . These inventories lead to the mean value of reference site in Augustenfelde in 2004 to be: 5417  $Bq/m^2$ .

In 2009 study, the result of measurements regarding reference sites, gave the values of 4666 Bq/m<sup>2</sup>, 4023 Bq/m<sup>2</sup> And 6456 Bq/m<sup>2</sup>. Taking the mean of three, the result will be, 5115.3 Bq/m<sup>2</sup>.

Next thing to consider is the information about the whole inventory of all 10 profiles. The inventory in upper 30 cm layer and the whole profile for all 10 study sites for 2004 study are shown in the following figure.



Figure 15. Cs-137 activities for all 1 to 9 eroding sites and the 10<sup>th</sup> depositional site (Kietzer, 2006) Purple bars: activity in the 30 cm upper layer (mBq/cm<sup>2</sup>) Yellow bars: activity in the whole vertical profile (mBq/cm<sup>2</sup>)

And the depth distribution of Cs-137 for profile 10 (depositional site) for 2004 study is shown in the following figure.



Figure16. Profile 10- depositional site, Cs-137 activities vs. Depth

There was no clear information about the density of each layer of soil in this study. Therefore, it was not possible to convert the activities to Becquerel per unit area  $(Bq/m^2)$  and compare it with the reference activities. Announced activities (Bq/kg) cannot really be used for comparison means, because the activity on each layer depends on the density (and thickness of each layer) which is not considered here.

Looking into details of the Cs-137 activities of profile 10, a sharp peak of 55-60 cm depth can be recognized. Above this layer there is almost a constant activity layer up to the surface and below it, a gradual decrease is recognizable.

The sampling for depositional site ended after 80 cm depth, therefore there is no information available about the Cs-137 activities below this layer.

There is no paper published on 2009 this study yet, but the results of measurements for all the 10 profiles are available.

Like the other study, first we will take a look at the results of the entire eroding sites, and then we will focus on the  $10^{th}$  profile (depositional site).

Like before, the inventory of upper 30 cm layer and the total inventory for each of the measurement sites are plotted.



Figure 17. Cs-137 activities for all 1 to 9 eroding sites and the 10<sup>th</sup> depositional site

The large difference between the inventory of profile 9 and the other eroding profiles is clear in this measurement as well as the former measurement on 2004. The  $10^{th}$  profile, depositional site, shows a very high inventory which is expected for a depositional site down the slope. Comparing the inventory of depositional site at 2004 and 2009, a very large increase is recognized, which is unlikely. The reason as it was mentioned before may be the difference in calculations of density. Focusing on the depositional site, the depth distribution of Cs-137 in profile 10 for 2009 study is shown in the following figure.



Figure18. Profile 10- depositional site, Cs-137 activities vs. Depth

As it is shown in figure 18, a somewhat constant Cs-137 activity is dominant in the upper 50 cm layer again. In this profile, like the  $10^{th}$  profile of the former study, a sharp peak can be recognized. In this case, the peak is located in 70 – 75 cm layer. Unfortunately the sampling stops on 80 cm and there is no information on inventory of the lower layers again.

#### 5.2. Measurement results of the current study

Sampling was performed at the same slope in Augustenfelde, focusing on the depositional site ( $10^{th}$  profile). Two vertical profiles were sampled on the depositional site. The sampling scale for both profile was the same and as shown in the following figure.

After all the preparation processes (explained in chapter 3), and Gamma-Spectrometry analysis, the activity of Cs-137 was measured for each layer.

The depth distribution of Cs-137 related to profile 10-1 and 10-2 in shown in the following plots.

a)



b)



Figure 19. Depth distribution of Cs-137 a) Profile 10-1 b) Profile 10-2

According to the measurement results and figure19, just like the other two vertical profiles of the depositional site, an almost constant Cs-137 activity is present in the upper 50 cm layer. But an obvious difference appears below this layer between profile 10.1 and 10.2.

Profile 10.1 shows a sharp peak at the 57.5-60 cm layer, similar to the profiles of the previous studies. But profile 10.2 not only does not include such a peak, but also shows an activity of almost zero between 50 and 60 cm depth. Measurement uncertainties are below 10% for all samples. Therefore, such a reduction in activities in profile 10.2 between 50 and 60 cm depth cannot be an effect of measurement uncertainties. It is possible that this reduction is a result of special variability of Cs-137 in soil due to some environmental disturbance or the sampling effect. In the following, the simulations and discussions are based on the profile 10.1.





Figure 20. Different layers of profile 10.1 defined according to the activity changes in each layer: constant activity, large peak, gradual decrease and small peak

In this profile, the large peak is located on 57.5-62.5 cm depth. Comparing it with the 2004<sup>th</sup> profile in which the same peak was located at 55-60 cm depth; a 5 cm difference can be recognized. Comparing the results with 2009<sup>th</sup> profile that had its peak on 70-75 cm depth we

can see that the peak is more than 10 cm lower than 2010 peak. Actually the peak of 2009<sup>th</sup> profile seems lower than where it is expected to be found.

In 2010<sup>th</sup> study there also exists a smaller peak at 90 cm depth. In previous studies, no information is available on the existence of such a maximum, because the sampling stopped at 80 cm in both 2009 and 2004 studies.

#### 6. Discussions

In this chapter, the improved depositional model (Jha & Kirchner, 2009) will be used to calculate the deposition rates. Then, this model will be applied with the three input functions that were introduced before (i.e. considering gradual weapons fallout, considering the Chernobyl atmospheric input, considering both of them simultaneously) and deposition rates will be calculated again. The results will be compared afterwards.

Then, the above mentioned modeling approaches will be fitted to the experimental data and the results will be discussed.

At the end, the results of the experimental data will be explained in details and the best simulation will be introduced and its advantages and disadvantages will be discussed.

#### 6.1. Deposition rate

Mathematical models are developed to predict the natural processes as close as possible to the reality. In this study, the natural process that is being studied is erosion and deposition and the erosion /deposition rate is the quantity that has to be predicted. Focusing on the depositional site (profile 10), the improved depositional model discussed in chapter 4 is used to find the deposition.

#### 6.1.1. Reference value of Cs-137 inventory

The reference value of the former study (Jha, 2009) was used for the calculations of erosion and deposition rates. The sampling of reference sites took place almost one year before this study, therefore in was corrected for decay to the reference date of sampling. The reference inventory used in this study is:  $5012 \pm 208$  Bq/m<sup>2</sup>

#### 6.1.2. Erosion rate

As mentioned in section 4.3, the first thing to be considered in deposition rate calculations, is the information about all the eroding sites.

In this case, the depositional site is located at the end of a moderate slope. It is expected that all the soil which is being eroded from the slope will accumulate at the depositional site. 9 profiles were sampled through the slope in the former study (Jha, 2009). Total inventories of these 9 profiles were used in estimations of erosion rate for each of the profiles. Their total inventories are shown in the following table.

Profile	Total Inventory (Ba/m <sup>2</sup> )
No.	
1	4093 ± 314
2	3828 ± 239
3	$4535 \pm 262$
4	4245 ± 227
5	$4038 \pm 174$
6	$4210 \pm 260$
7	3918 ± 194
8	2124 ± 178
9	538 ± 80

Table2. Total inventory of the 9 eroding profiles

Using the simplified mass balance model of equation 6, reference inventory and the total inventories, the erosion rate can be calculated for each of the 9 profiles.

The erosion rates, calculated for both models are as follows.

Profile	Erosion rate (cm/yr)	Erosion rate (cm/yr)
No.	Fallout in 1963	Gradual fallout
1	0.14	0.13
2	0.19	0.17
3	0.08	0.07
4	0.12	0.11
5	0.16	0.14
6	0.13	0.12
7	0.16	0.15
8	0.49	0.44
9	1.36	1.23

Table3. Erosion rates of the 9 eroding sites

The reason for the slight difference between fallout in 1963 and gradual fallout forms is because of the difference between starting value of time.

It has to be mentioned that all models only give long term mean erosion rates and it is not possible to estimate the variability of erosion rates using these models.

#### 6.1.3. Eroded Cs-137 concentration

The Cs-137 concentrations eroded from each of the eroding sites are the next quantities to be calculated using equation 8.

Rearranging the simplified mass balance model, using erosion rates calculated in the previous section and reference inventory of each year (from the beginning of atmospheric fallout up to the sampling date), the concentration of Cs-137 eroded each year, from each of the profiles can be estimated.

Using each of the models, the eroded concentration through time for each of the 9 sampling points will be different.

#### 6.1.4. Deposited concentration

The deposited depends on eroded concentrations, erosion rates and the area of each eroding site.

$$C_{d}(t_{l}) = \frac{\sum_{i=1}^{k} C_{e_{i}}(t_{l}) . \Delta Z_{i} . S_{i}}{\sum_{i=1}^{k} \Delta Z_{i} . S_{i}}$$
(15)

Where, k is the number of eroding sites and  $S_i$  is the area of each site.

The rest of the terms are the same as before.

To calculate the area of each sampling point, the lengths of the slopes between each profile were calculated. 30 meter width was assumed for all the profiles. The result of multiplication of length and width, gives the area of each profile.

Profile	Area
No.	(m <sup>2</sup> )
1	720
2	750
3	750
4	750
5	780
6	750
7	570
8	450
9	600
10	600

Table4.Area of each profile

Deposited concentration that is the weighted eroded concentrations of all the eroding sites was estimated through time using both original and gradual model, using equation (15).

 $C_d(t)$  is still not the Cs-137 concentration of the depositional site through time. It is just the concentration of Cs-137 in the soil carried to the depositional site. Which then is modified by taking into account the effect of plowing, concentration of Cs-137 at the depositional site can be estimated using the improved depositional model.

$$C_{dep}(t) = \frac{C_d(t - \Delta t).(d - \Delta Z) + C_d(t).\Delta Z.e^{-\lambda(t - t_0)}}{d}$$
(16)

All the terms are the same as before; the only parameter which needs discussion is  $\Delta Z$ .

 $\Delta Z$  is usually considered as the sum of all the erosion rates, or the net rate, including all the eroding and depositional sites. But the areas of the eroding and depositional site are not included in these approaches. In this study a different approach is used for the calculation of  $\Delta Z$  which is described in the following.

Let's look at the erosion event on the study site from a slow motion camera. Erosion starts at the top of the slope (profile 1). A soil layer of  $\Delta Z_1$  is being eroded from the area of profile 1 and reaches the second point. Looking at the second point at this moment, it looks like a deposition site. The soil which is washed from the area of profile 1 will spread over the area

of profile 2. One moment later, all the soil carried away from profile 1 is summed up with some soil from profile 2 and rushes to the area of profile 3, which is the one experiencing deposition the next moment. This process will continue along the slope and will stop at the end of it. The amount of soil that reaches the depositional point will spread over its area.

Therefore, Area of each profile  $\times$  erosion rate of that profile is the volume of soil deposited over the area of the next profile. This deposited layer, plus the eroded soil of the second point is the layer which enters the third section. Travelling, point by point with the soil that passes through each profile, the thickness of the layer that will accumulate over the area of the last profile can be estimated as follows.

Profile  $1 \rightarrow \Delta Z_1 \times S_1 = \Delta Z_1 \times S_2$ Profile  $2 \rightarrow \Delta Z_2 + \Delta Z_1 = \Delta Z_{net_2}$ ,  $\Delta Z_{net_2} \times S_2 = \Delta Z_2 \times S_3$ Profile  $3 \rightarrow \Delta Z_3 + \Delta Z_2 = \Delta Z_{net_3}$ ,  $\Delta Z_{net_3} \times S_3 = \Delta Z_3 \times S_4$ 

Profile 9  $\rightarrow \Delta Z_9 + \Delta Z_8 = \Delta Z_{net_9}$ ,  $\Delta Z_{net_9} \times S_9 = \Delta Z_9 \times S_{10}$ Profile 10  $\rightarrow 0 + \Delta Z_9 = \Delta Z_{net_{10}}$ 

Using this approach  $\Delta Z_{net_{10}}$  is calculated both using 1963 fallout and gradual fallout deposition functions. Results are shown in the following table.

Simplified Mass	$\Delta Z_{net}$
<b>Balance Model</b>	
1963 fallout	2.9 cm/yr
Gradual fallout	2.6 cm/yr

Table5.  $\Delta Z_{net}$  calculated with both 1963 fallout and gradual fallout form of simplified mass balance model This calculations demonstrate that,  $\Delta Z_{net}$  depends strongly on the area of the depositional site and this dependency and also neglecting the effect of plowing may bias the results.

Going back to the calculation of deposition rate using improved depositional model, deposited concentrations of each year can be calculated. Though using the gradual form of

this model, one extra term which describes the atmospheric input of each year has to be added to the equation.

$$C_{dep}(t) = \frac{C_d(t - \Delta t).(d - \Delta Z) + C_d(t).\Delta Z.e^{-\lambda(t - t_0)} + I(t)}{d}$$
(17)

I(t), is the atmospheric input in term of inventory(Bq/m<sup>2</sup>) related to each year; from 1958 to 1981 there are inputs of weapons fallout and on 1986 there is the input of Chernobyl accident.

#### 6.1.5. Deposition rate

Using the quantities estimated in the previous sections deposition rates are calculated (equation 16 and 17).

Deposition rates which were estimated using the 1963 fallout function and other three form of it which were used before (gradual weapons fallout, including Chernobyl and considering both simultaneously), are shown in the following table.

For these simulations it was assumed that Chernobyl derived Cs-137 is 50% of the weapons fallout Cs-137 input on 1986 and weapons fallout was calculated using the reference profile in a way that reference value for today will be,  $5012 \text{ Bq/m}^2$ .

Improved	depositional	Deposition rate (cm/yr)
model		
1963 fallout		0.90
Gradual weapons fallout		0.93
1963 fallout + Chernobyl		1.39
Gradual fallout + Chernobyl		1.10

Table6.Deposition rate calculated using different forms of the improved depositional model

It can be recognized that the deposition rate in underestimated by the original model and once again the importance of considering the Chernobyl input on 1986 becomes apparent. The difference between the results of 1963 fallout form with and without Chernobyl is clearly visible; considering the Chernobyl input results in a 40% increase in deposition rate. Considering both gradual fallout and the Chernobyl accident input will moderately affect the simulations resulting in a 20% difference between the 1963 fallout form and the one which considers both gradual fallout and Chernobyl input.

#### **6.2.** Fitting processes

One of the most common methods to check whether the model which is chosen to describe a natural event is suitable or not, is to fit the free parameters of it to the experimental data and to compare its predictions using these adopted parameters with independent data. It means that the predicted values of any quantity will be compared with experimental data while the final goal is to minimize the difference of these two data sets. This can be done by changing the value of free parameters in the model and by fixing them to the values which give the least deviations from the measurements.

In this study, the Cs-137 concentrations calculated by the improved depositional model are going to be fitted to the measurement results of the 10<sup>th</sup> profile (depositional site).

The 1963 fallout model is not a complete description of the real situation. It can be used to approximate the deposition rate but to reconstruct the vertical profile more detailed forms of the model are preferred. Therefore most of the fitting processes were applied on the most detailed form which includes the gradual weapons fallout and Chernobyl accident inputs.

#### 6.2.1. Extracting the information from the experimental data

The measurement results of the depositional site, which has been sampled for this study, were presented before. To be able to use these data in fitting, they have to be processed and converted to a quantity that can be compared with the model outputs.

The model gives the concentration of Cs-137 over time  $C_{dep}$  (Bq/m<sup>3</sup>) at the depositional site.

These concentrations can provide the depth distribution of Cs-137 in soil, when the deposition rate is known.

On the other hand, measurement results are in terms of specific activity (Bq/kg). These activities have to be converted to concentrations (Bq/m<sup>3</sup>) to enable a comparison between model products and measurement results. This can be done by multiplying the specific activities of each sampled layer by its density which has been calculated for each layer in the sample preparation process.

A table of measured Cs-137 activities, densities and converted values is presented in the following.

The uncertainties of densities were so low and negligible.

Depth (cm)	Specific activity (Bq/kg)	Density (kg/m <sup>3</sup> )	Concentration (Bq/m <sup>3</sup> )
0 - 5	$6.40 \pm 0.23$	1510	9664 ± 347
5 - 10	5.81 ± 0.23	1630	$9470 \pm 375$
10 -15	$5.75 \pm 0.30$	1550	8913 ± 465
15 - 20	$6.31 \pm 0.38$	1790	$11295 \pm 680$
20 - 25	5.10 ± 0.39	1150	$5865 \pm 448$
25 - 30	5.37 ± 0.48	1600	8592 ± 768
30 - 35	5.76 ± 0.32	1620	9331 ± 518
35 - 40	$4.83 \pm 0.38$	1670	$8066 \pm 634$
40 - 45	$4.04 \pm 0.34$	2120	8565 ± 720
45 - 50	$5.12 \pm 0.46$	1940	9932 ± 892
50 - 52.5	$7.76 \pm 0.37$	1740	$13502 \pm 644$
52.5 - 55	8.45 ± 0.46	2240	$18928 \pm 1030$
55 - 57.5	$10.1 \pm 0.6$	1840	$18584 \pm 1104$
57.5 - 60	50.8 ± 1.9	1330	67564 ± 2527
60 - 62.5	$23.7 \pm 0.9$	1060	25122 ± 952
62.5 - 65	$15.5 \pm 0.7$	1140	$17670 \pm 798$
65 - 67.5	$10.5 \pm 0.7$	1340	$14070 \pm 938$
67.5 - 70	8.10 ± 0.30	1010	8181 ± 303
70 - 72.5	7.44 ± 0.53	1570	$11681 \pm 630$
72.5 - 75	$5.89 \pm 0.58$	1190	$7009 \pm 690$
75 - 77.5	$3.72 \pm 0.27$	1150	4278 ± 310
77.5 - 80	$1.68 \pm 0.30$	1310	2201 ± 393
80 - 82.5	$1.00 \pm 0.27$	1260	$1260 \pm 340$
82.5 - 85	$0.82 \pm 0.09$	1140	$935 \pm 102$
85 - 87.5	$1.61 \pm 0.21$	1270	$2045 \pm 266$
87.5 - 90	$2.55 \pm 0.46$	1270	3239 ± 584
90 - 92.5	$0.93 \pm 0.28$	1280	$1190 \pm 358$
92.5 - 95	$0.68 \pm 0.17$	1340	911 ± 227
95 - 97.5	0	1040	0
97.5 - 100	0	1000	0

Table7. Measured value of Cs-137 specific activity, density and concentration for each sampled layer

#### 6.2.2. Parameter estimation

Other than measured quantities, there are some parameters such as the plough depth, total Cs-137 atmospheric input of nuclear weapon tests and Chernobyl Cs-137 input which are used in the model and have to be estimated from the existing records. In some cases the fitting method can be used to find the best value of each parameter as well.

In the following a brief review of the records and documents for each parameter will be presented.

Plough depth has been studied in both of the former studies and was estimated for all the sampling profiles. It may change through the field, but not drastically. In most of the cases in Augustenfelde, the plough depth was 25 to 35 cm. as a result a mean plow depth of 30 cm was used in the calculations. It has to be mentioned that plow depth can be different for each sampling area, due to the method and machinery used for the plowing processes.

The next parameter to be estimated is the total Cs-137 atmospheric deposition of nuclear weapon tests. There are different records of Cs-137 weapons fallout plus Chernobyl input but now it is not possible to separate them anymore. At the time of the accident Cs-134 was also presented in the fallout. The ratio between the production of Cs-134 and Cs-137 could be estimated and therefore the amount of Cs-137 fallout of the Chernobyl accident was known. But, due to the decay of Cs-134 (2 years half life) weapons fallout and Chernobyl fallout can no longer be separated.

The most reliable reports about the atomic radiations have been published by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation).

Referring to the UNSCEAR report (Sources and Effects of Ionizing Radiation, Volume1: Sources, United Nations, 2000), the total Cs-137 nuclear weapon tests deposition for the 50 - 60 degree latitude band, was estimated as **3900** Bq/m<sup>2</sup>.

As I mentioned, the Chernobyl accident's Cs-137 atmospheric input cannot be estimated independently. There are some records about the summation of nuclear weapon tests and Chernobyl depositions in different publications but the suggested values encompass wide ranges from 2000 to 10000 Bq/m<sup>2</sup>. Therefore another approach was used here to estimate Chernobyl's Cs-137 atmospheric input. In this approach, the reference inventory of sampling date is back decayed to 1986 (Chernobyl accident year), at the same time the weapons fallout which has its last input on 1981 (based on the profile used in this study) was decayed corrected to 1986, the difference between these two values indicated the Chernobyl share on 1986. Using this method, the Chernobyl accident's Cs-137 deposition was estimated as **6125** Bq/m<sup>2</sup>.

It is obvious that estimations of weapons fallout and Chernobyl accident Cs-137 input can only be used in the most detailed form of the model which includes both Chernobyl and gradual bomb testing atmospheric input. Applying other form of the improved depositional model (original or gradual), the back decay method (explained before) was used. For example, back decay calculations for the original model that have only one Cs-137 input, gives the value of about 14000 Bq/m<sup>2</sup> atmospheric deposition on 1963 which is extremely higher than the expected values.

#### 6.2.3. Deposition rate estimations

Using the parameters estimated in the previous section in the detailed form of the improved depositional model to calculate the deposited concentrations over time  $(C_{dep})$ , and then inserting them in equation 19 to find the deposition rate, the following result was derived.

Improved depositional	Deposition rate	
model	(cm/yr)	
Gradual + Chernobyl form	1.48	

Table8. Deposition rate calculated using best values of the parameters

Then, the calculated concentrations  $(C_{dep})$  were fitted to the experimental data to estimate the deposition rate and its uncertainties.

Fitting in this case is not a straight forward processes. Layers of experimental data are not uniform and obviously more than one annual deposition corresponds to each sampling layer.

A model was developed in MATLAB. This model let the deposition rate to change freely until the deposition rate which minimizes the deviation between experimental data and calculated concentrations is found. The non linear regression was used as the fitting method. The result of fitting was a deposition rate of  $1.38 \pm 0.09$  cm/yr.

The difference between the best estimate of deposition rate and the one which is calculated by directly using the model is less than 10%.

The plot of calculated concentrations in depth and experimental data, while the deposition rate is set to 1.38 cm/yr is as follows.



Figure21.Cs-137 concentration vs. depth (calculated by the model - experimental data)

Fitted concentrations are considerably close to the experimental data, with the exception of the sharp peak in 57.5-60 cm depth. In the 10 cm below the peak a different behavior is visible between the measured values and the theoretical model.

The sharp peak was also detected in the prior studies at the depositional sites. A potential explanation about the formation of such a peak was based on the soil texture and particle size change in that depth (Kietzer, 2006).

This reasoning is not convincing. As it is clearly shown in figure21, such a peak makes a great change in total inventory of the profile. If the formation of peak was due to the transport of Cs-137 from upper layers to lowers and trapping in a specific layer, a decrease in activity of upper layers would have been obvious. However, comparing the activities in upper layers of depositional site and all the eroding sites such a decrease is not visible.

A more plausible explanation is based on the fact that some of the Cs-137, which was deposited through the slope due to the atmospheric fallout, may be transported down the slope freshly before the annual plowing.

Erosion is a gradual and continues process and it is not easy to determine the dislocation of each of the erosion events but obviously the amount of soil which was eroded immediately after the fallout contains much more Cs-137 than the soil which is being transported after plaguing.

On the other hand, atmospheric fallout, deposition of the nuclear weapon tests fallout, also was a gradual process. Therefore the only event in which a high deposition at a single time occurred was the Chernobyl accident. As a result the soil which has been eroded immediately after the atmospheric deposition of Chernobyl accident contains a very high amount of Cs-137 and could contribute to an accumulation of high activities at depositional sites shortly after the accident.

The depositional site (profile 10) is placed at the end of a moderate slope in which 9 eroding sites has been studied. If some percentage of the Chernobyl accident's Cs-137 was eroded before the plowing, the inventory of all the eroding sites will be a little bit smaller than the expected values, whereas these small activities will accumulate on the depositional site down the slope, at the 10<sup>th</sup> profile.

In order to find the best value for the amount of soil transported with this process and to check how the deposition rate will differ when applying such a change, the fitting method was used again. In this fitting process, an unknown percentage of Cs-137 was removed from all eroding sites and the same percentage from each of the eroding sites was added to the depositional site. The percentage and corresponding deposition rate are free parameters and the best estimate is the one that fits the experimental data the best.

The results give a  $34\% \pm 6\%$  reduction of activity on eroding sites in 1986 while the deposition rate reduces slightly to  $1.31 \pm 0.02$  cm/yr. The simulated concentrations using these parameter values are plotted in the following figure.



Figure 22. Cs-137 concentration vs. depth (calculated by the model – experimental data) After applying the changes

Now that the high peak at 57.5-60 cm depth and the smaller one on 87.5-90 cm depth are reproduced. Major discrepancies still are present in the area below the high peak. In this area the activities predicted by the model are much lower than the measured ones.

This indicates that there are some physical processes that are not included in the model. The downward diffusion and convection are physical processes which may produce high activities below the peak. This also is the reason that in reference sites that were not the subject of erosion, deposition and plowing, Cs-137 can be found at least down to 20 cm depth. Referring to the reference sites that have been studied for this study, the Cs-137 has been transported 15 to 20 cm down by diffusion and convection. Because the 10<sup>th</sup> profile is located in the same area, a soil texture similar to reference is expected. Therefore Cs-137 can be expected to be transferred by convection and diffusion at both sites similarly.

The difference between remnant of nuclear weapons fallout and the measured concentrations between 60 and 80 cm depth were the Cs-137 inventory which was transported downward due to diffusion (mainly) and convection.

The same fitting processes can be applied to the gradual fallout and only Chernobyl forms of the improved depositional model to quantify their consequences.

Assuming that the only atmospheric input of Cs-137 was the nuclear weapons tests in its gradual form, the results will be a total Cs-137 input of almost  $17000 \pm 3000$  Bq/m<sup>2</sup> and the deposition rate of 0.78  $\pm$  0.05 cm/yr.



Figure23. Cs-137 concentration vs. depth (calculated by the model – experimental data) Improved depositional model considering only gradual weapons fallout

Because the Chernobyl accident is not included all the information about the existence of two peaks is lost. This version of the model cannot describe the measured profile correctly. In addition, large value suggested for the total atmospheric input of nuclear weapon tests is not reasonable at all.

The same procedure was repeated while only the Chernobyl accident was considered. Therefore the measured data below the peak was not considered in the fitting process. The resulting Chernobyl input was about  $9700 \pm 1000$  Bq/m<sup>2</sup> with a deposition rate of  $1.22 \pm 0.05$  cm/yr.



Figure24. Cs-137 concentration vs. depth (calculated by the model – experimental data) Improved depositional model considering only Chernobyl

Taking into account the Chernobyl input and ignoring the weapons fallout will cause the information below the peak to be lost. The advantage of this fit is that it gives the peak exactly where it is expected.

#### 6.2.4. Testing the best fit with previous measurement results

Now that the best fit for the measurement results of this study is found, it has to be tested with other depositional site measurement results to check its validity.

In the following the measurement results of the last year's sampling (Jha, 2009) is compared with the best fit.



Figure 25. Cs-137 concentration vs. depth Calculated by the model (best fit) – experimental data (2009)

While using the best fit of the improved depositional model, the inventories and the reference values have been decay corrected to the date of the sampling (2009).

As it is shown in figure 25 the best fit predicts the sharp peak perfectly, though it is not exactly at the same depth as the measured peak. It has to be mentioned that some part of this 5 cm difference may be due to the systematic errors of sampling and the uncertainties of the model itself, but even with this deviation, our model still is a good approximation of the real depth distribution.

The other measurement that took place at the same area was by Kietzer in 2004. In the following the measurement results of this sampling are compared with the best fit as well. The information about the soil density of each layer was not well documented and the total inventory calculated for this profile seems to be somewhat low.



Figure26. Cs-137 concentration vs. depth Calculated by the model (best fit) – experimental data (2004)

The reference value and total inventories for eroding sites of the 2004 study were used in this comparison. As it is shown in figure 26 the location of the peak is predicted perfectly although some difference for the 1986 peak is present.

#### 6.2. Error analysis

Analyzing the uncertainties is a very important part of any estimation and the error propagation method is a very common way to do it. In this method, the uncertainties of variables of a function are used to find the uncertainty of the function itself. In this study, uncertainties of the results of experimental measurements are due to the measurement limitations and human errors. Uncertainties of the measurement equipments are known and human errors can be estimated approximately depending on the experiment.

On the other hand, in this study for estimations of best deposition rate the fitting method was used. In this method the estimated value is always accompanied with the standard error of it (which was documented for all the results). The uncertainty of measurements was presented before.

#### **6.3.** Conclusion

The simulations and fitting processes show that in general the improved depositional model in its original form can be used to predict the deposition rate and Cs-137 depth profile, though it does not give a detailed explanation of the real case and the final results deviate about 35% from actual data. Specifying the atmospheric deposition of the weapons fallout and the Chernobyl ratio explicitly give much better results and are more successful in predicting the vertical profile in details.

Our experimental data show that other than regular atmospheric fallout, erosion, plowing and deposition some other natural processes may effect the concentrations of Cs-137 in soil. Taking into account some of these processes (in their simplest form) made the model predictions almost identical to the experimental data.

One of the most important events which affect the depth profile of Cs-137 is the fact that some of the Cs-137 originating from the Chernobyl accident may have been transported to the depositional site freshly before the plowing. The immediate transportation of small amounts of soil which includes high activities produced a sharp peak at the year of the accident in the Cs-137 profile. Afterwards, convection and diffusion transported some of this Cs-137 downward.

With consideration of these processes it was possible to reconstruct the real profile almost perfectly. The estimated deposition rate and the shape of the predicted profile matched the other studies' measurements results with a good approximation as well.

#### **Summary**

Erosion and sedimentation processes on agricultural fields have been a major concern and problem around the world for the last few decades. Many studies have been dedicated to this subject and many papers have been published about different ways to estimate and predict it. One of the most efficient methods is to use Cs-137 as a tracer of erosion or deposition. Various mathematical models have been developed for this method to enable us to estimate the rates of erosion/deposition accurately. One of these models which is designed especially for the studies of depositional sites is the improved depositional model and was introduced by Jha and Kirchenr (2009).

The goal of this study was to validate this model, to estimate its precision and if necessary to improve this model.

In order to reach this goal, concentrations of Cs-137 in soil were measured by sampling the vertical layers of soil and measuring the Cs-137 inventory in each layer. Then, the model was used to reproduce the measurements, the results were compared and the differences were recorded. Afterwards, in order to get one step closer to the reality, some simplifying assumptions were removed from the model; first, the assumptions which consider only a single atmospheric Cs-137 deposition event in 1963 and second the assumption not to include Chernobyl in the calculations. After removing these assumptions, a detailed form of the model was established and the calculations of Cs-137 depth distribution were repeated. The results were compared with the original model and the experimental data and the improvement of the model was recorded. Next step was to include some more natural processes (In particular, erosion events which occur before the plowing and diffusion) in the model. This change gave predictions of vertical profile of Cs-137 which could reproduce the experimental data very well. Its performance was further checked with the detailed form of the model is able to predict deposition rates of soil mobilized by erosion accurately.

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