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Evaluation of spatial variability in comparative toxicity potential of copper in soils for application in life cycle assessment

Master thesis

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Miskolc, 2013. Május 8. ...................................................

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III. ABSTRACT

Soil properties show significant spatial variability at local, regional and continental scales. This is a challenge for life cycle impact assessment (LCIA) of metals, because fate, bioaccessibility, bioavailability, and effect factors are controlled by environmental chemistry and can vary orders of magnitudes for different soils. The aim of this project is to evaluate spatial variability in the terrestrial ecotoxicity characterization factor (CF) or comparative toxicity potential (CTP) of copper for application in regionalized and global-generic life cycle assessment.

I calculated CTPs of copper in soils after unit emission to air for each of 1x1 km grid cells of the Harmonized World Soil Database (HWSD), using regression equation presented in (Owsianiak et al 2013). Then deposition fields (independent of emission source) for the soil in each of the grid cells using different atmospheric deposition models has been created and evaluated then based on the previous steps I developed weighting schemes to calculate emission-specific CTP of Cu.
A szálló por réztartalom összehasonlító toxicitási potenciáljának térbeli változása a talajban és értékelése az életciklus-vizsgálatban való felhasználás céljából

A különböző légszennyező szálló porokat bocsátó pontforrások nagy mennyiségű szilárd anyaghoz kötöt fémet, köztük rezet is bocsátanak ki. Ezek ülepedése a pontforrás környezetében lévő talajok rézkoncentrációját növeli. Az életciklus vizsgálat során a különböző kibocsátott toxikus anyagokhoz ún. összehasonlító toxicitási potenciált rendelnek, annak érdekében, hogy a teljes toxicitást fel lehessen mérni és össze lehessen hasonlítani, akár különböző anyagok esetén is.

Egy adott talajban, annak tulajdonságaitól nagy mértékben függ, hogy a mérgező anyag végül is valóban ki tudja-e fejteni hatását, vagy kötött, inaktív állapotban marad. Ezek a talajtulajdonságok nagymértékű változékonyságot mutatnak a földrajzi térben, ezért az életciklus vizsgálat szempontjából lényeges, hogy figyelembe vegyünk ezeket a térbeli mintákat. Nem mindegy, hogy 1 kg szálló porhoz kötött rezet a földgolyó mely pontján eresszük szélnek, ugyanis az ülepedő porból a talajba kerülve igen eltérő módon fejtheti ki hatását függően a fogadó közeg érzékenységétől.

Az életciklus-vizsgálatban eddig ezeket a térbeli változásokat nagyrészt figyelmen kívül hagyó modelleket használtak a különböző fémek toxicitási potenciáljának kifejezésére. Jelen dolgozatban ezért egy olyan módszert mutatok be, amely légköri terjedési és ülepedési modellek segítségével figyelembe veszi az említett térbeli különbségeket, és 1 egység réz toxicitási potenciálját aszerint súlyozza, hogy melyik érzékenységű talajra mennyi egység ülepedik ki. Mindehhez a magyar széntüzelésű erőművek esetét dolgoztam ki, így végül egy országra vetített átlagos toxicitási potenciált is kidolgoztam, amely kifejezi, hogy 1 egység magyar hőerőművekből kibocsátott réznek mekkora átlagos toxicitási potenciálja lehet.
CHAPTER 1: INTRODUCTION

1.1 STRUCTURE OF THE THESIS

In the first chapter a general overview of life-cycle assessment (LCA) is presented with special regard to spatial and geographic differentiation in the context of terrestrial ecotoxicity assessment of metals. Then problem formulation is provided along with the end goal of this study. The second chapter describes and evaluates the different methods, tools and datasets which is used in this thesis to provide solutions for the problem. After presenting the results in Chapter 3, a discussion of conclusions follows in the last Chapter. The Appendix contains the technical details of numerical modeling and some data tables containing values of important parameters obtained from external sources.

1.2 CONTEXT

1.2.1 Life-cycle Assessment (LCA)

Environmental sustainability is surely one key issue for the humankind to survive the 21st century. In order to achieve more sustainable production and consumption, a number of tools and methods has already been developed to identify and better understand the environmental impacts of production of goods and services, e.g. environmental impact assessment (EIA), ecological footprint, life-cycle assessment or Design for Environment (DfE).

From the late 1960’s a new approach has been emerged to investigate the environmental consequences of goods throughout their production chain. Starting from early simplistic mass balances and gate-to-gate approaches, the method gradually became more and more complex, and today life-cycle assessment (LCA) methodology reflects on several environmental issues applying the concept of life-cycle viewpoint (Guinee & Heijungs 2010).

It quantifies and evaluate the potential environmental and health impacts associated with any good in its all stages of life-cycle from resource extraction, through production, trade, transport, use, recycling, to the end of life or disposal. Usually LCA looks at products from
a functional viewpoint and considers all things that are needed to provide a given function. For example the function of providing text on a sheet of paper for reading needs wires, electricity and a printer with ink and paper in it. However LCA goes deeper to the production chain from extracting the fossil fuels for electricity generation, to the use of wood and water for papermills etc.

Moreover, the holistic view of LCA helps to avoid reducing one environmental impact while creating an other, the so-called “shifting of burdens”.

From the early 1990's the Society of Environmental Toxicology and Chemistry (SETAC) has had the leading role in the development of the science behind the LCA methods by organising workshops and establishing LCA Advisory Group for experts. In parallel a standardisation process has initiated by the International Standardisation Organisation (ISO) in an effort to harmonize local and national environmental standards. In 1997 ISO published the first version of LCA standards (ISO14040 and 14044) in the framework of the ISO14000 series (Environmental Management Standards).

Thus, LCA is an important sustainability tool to support decision making based on environmental performance of products and an effective way to look at the environmental hot-spots throughout the production, including supply chain and downstream (ILCD 2010)

1.2.1.1 The phases of LCA

According to the ISO14044 standard, four phases have to be included in an LCA study:

- goal and scope definition,
- inventory analysis,
- impact assessment, and
- interpretation.

In the goal and scope phase the system boundary and the level of detail is (scope) as well as the intended use and applications (goal) are defined. The goal of a given LCA greatly determines the depth of analysis, e.g. which unit processes are included in the system. The system boundary clarifies not only the life-cycle stages but the interfaces where the exchanges occur between the technological system and the ecosphere.
In the second, life-cycle inventory (LCI) phase, an inventory of all elementary flows (substance flows which cross the boundary between human activity and the environment) are collected in line with the system being studied. The first LCA studies consisted of only an inventory of different substances summed up for the whole life-cycle.

**Figure 1. Phases of an LCA (ISO14040:2006)**

In the second, life-cycle inventory (LCI) phase, an inventory of all elementary flows (substance flows which cross the boundary between human activity and the environment) are collected in line with the system being studied. The first LCA studies consisted of only an inventory of different substances summed up for the whole life-cycle.

**Figure 2. The scheme of system boundaries in LCA**
For example in the case of electricity production in coal-fired power plants, there are a big number of different substances are emitted, contributing to different environmental problems with varying magnitude. From water vapor and carbon-dioxide (causing global warming) to acidifying sulphur-dioxide and particle-bound metals (that are toxic to soil), there is a wide range of substances and possible effects.

The life cycle impact assessment phase (LCIA) is the third phase of the LCA. From the ISO standard: “The purpose of LCIA is to provide additional information to help assess a product system’s LCI results so as to better understand their environmental significance.” (ISO 14044 2008). In practice, in the LCIA phase, the inventoried substances (such as water vapor, carbon-dioxide or airborne metals) are classified into a limited number of environmental problems (or impact categories) and their relative contribution is assessed in order to create aggregated indicators. In other words, during the LCIA procedure the environmental interventions (emissions and resource depletion) are assessed in terms of contribution to the different environmental impact categories, such as human toxicity, climate change, eutrophication etc.

The two obligatory key steps of LCIA are classification (assignment of each substance in the inventory to one or more impact categories) and characterisation (conversion of substance flows to impacts using characterisation factors). Characterisation factors (CFs, or in terrestrial ecotoxicity assessment comparative toxicity potentials - CTPs) represent the contribution from an elementary flow to an impact, and for a given environmental category the impact score can be obtained by multiplying the amount of \( n \)th substance in the inventory in its relevant unit (\( m_n \)) with its relevant characterisation factor (\( CF_n \)):

\[
impact \ score = \sum_{i=1}^{n} m_i \cdot CF_i \quad (1)
\]

The impact score is often represented as an equivalent of a given reference substance (e.g. kg CO\(_2\) equivalent in the case of climate change, see Figure 3 for an example). Calculation of characterisation factors are always based on modelling of the environmental mechanism.
Basicly, the results of a LCIA can be seen as an environmental profile, where elementary flow substances are grouped in a limited number of impact category.

Finally in the interpretation phase the significant issues are identified as well as recommendations, limitations and uncertainty are evaluated and reported.

### 1.2.2 LCIA of metals in terrestrial ecotoxicity assessment

Among all impact category, terrestrial ecotoxicity is the one that probably needs most further development in terms of characterisation model. Even The International Reference Life Cycle Data System (ILCD) Handbook which is aimed at LCA practitioners, does not provide a recommended method for it, due to the lack of scientific consensus and poorly developed existing methods (Hauschild and Goedkoop 2013).

In ecotoxicity impact category following (Gandhi & Diamond 2010), all relevant emitted substance in the inventory has a comparative characterisation factor (CTP or characterisation factor - CF) which are "quantitative estimates of the ecotoxicological
impacts of substances per unit emission via pathways of exposure to defined environmental recipients”. That means the translation of emissions into potential adverse effects, in our case into Potentially Affected Fraction (PAF) of species in the given compartment. “The PAF can be interpreted as the fraction of species that is exposed to a concentration equal to or higher that the no observed effect concentration” (Goedkoop & Spriensma 2001) in relation to the unit emission of the toxic materials. This is very useful if we want to compare the toxic effects of e.g. 1 kg airborne copper to 1 kg airborne cadmium in the soil. In ecotoxicity, comparative toxicity potentials (CTPs or characterisation factors - CFs), are often differentiated by the receiving compartments (Rosenbaum & Bachmann 2008). That means that e.g. 1 kg emitted copper will have different CTPs (or CFs) if it’s receiving environment is soil or freshwater or marine environment. This approach is a very big step towards fair assessment, but has it’s own drawbacks, because it does not take into account the high variability of sensitivity between soil types (Owsianiak et al. 2013).

For airborne metals, generally the amount of emitted mass will deposit relatively closer to the emission source (e.g. industrial stacks) with higher probability, but other factors such as particle properties (size distribution, density, shape etc.), meteorological conditions (wind speed, turbulence, precipitation etc.), emission source (stack height, outlet pressure and temperature etc.) and terrain surface type might modify the deposition pattern. Moreover, small particles (below 1 μm size) can travel very far away, even thousands of kms (Chen & Stein 2013).

Then the toxicity potential of the deposited metals depends on the accessible and bioavailable fraction of the specific metal, and on the fate, or in other words, on the residence time that it spends in soil in available form for the organisms (see Figure 4.).

The calculation of CTP in freshwater ecotoxicity presented by (Gandhi & Diamond 2010) and further modified for application in terrestrial ecotoxicity by Owsianiak et al. (2013). The authors introduced an accessibility factor (ACF):

$$CTP_{i,s} = FF_{i,s} \cdot ACF_s \cdot BF_s \cdot EF_s$$  (1)

where $CTP_{i,s}$ ($m^3/kg_{total emitted to air} \cdot day$) is the comparative toxicity potential of total metal s emitted to compartment $i$; $FF_{i,s}$ (day) is the fate factor calculated for total metal s in soil;
ACF_s (kg\_reactive/kg\_total) is the accessibility factor defined as the reactive fraction of total metal s in soil; BF_s (kg\_free/kg\_reactive) is the bioavailability factor defined as the free ion fraction of the reactive metal s in soil; and EF_s (m^3/kg\_free) is the terrestrial ecotoxicity effect factor defined as PAF for the free ion form of the metal.

Soil properties play big a role in all four factors. E.g. the availability of the toxic forms of metals is determined by the sorption of metals to soil colloids such as clay and organic carbon, while the base cations and protons can decrease the toxic effects.

1.2.3 Regionalization in LCA and geographic implications

The scope of LCA is global, however a number of factors, which influence the LCA results, can vary in geographic space (e.g. resources, climate, biosphere or even cultural settings.) Those geographical and spatial differences can highly determine the environmental performance of production. E.g. producing 1 kWh electricity in Hungary can be very different from producing it in Norway if we look at the resources and the emissions. But even if the same substances are released, the impacts can differ due to differences in climate, soil conditions, population density, etc. Somehow it needs to be
addressed in LCA to avoid false results.

First Potting & Hauschild (2004) emphasized the importance of role of spatial differentiation in LCA. On the one hand the industrial production along with its resources needed and emissions generated takes place in different locations all over the globe. This problem needs to be addressed in the inventory, and collecting data suffers from the general data availability problems. Usually it is already implemented in some ways (in country-level) but it has still a lot to develop. On the other hand unit environmental intervention causes varying impacts throughout space (Mutel et al. 2011). This comes up in the impact assessment phase (Finnveden et al. 2009).

In LCIA each impact category has different level of spatial detail is necessary depending on the underlying natural phenomena. E.g. climate change is almost insensitive to where the emission has occurred, but e.g. eutrophication or acidification is much more spatially dependent impact category. To cope with it, a distinction has been made between site-generic (globally relevant), site-dependent (regionally applicable) and site-specific (only locally valid) impact assessment (Potting & Hauschild 2004). Recent methods include some kind of spatial detail already with the division of emissions by compartment (Heijungs 2012). E.g. emissions to air normally were treated differently from emissions to groundwater.

Table 1: Spatial differentiation in different impact categories (From: (Potting & Hauschild 2004))

<table>
<thead>
<tr>
<th>IMPACT CATEGORY</th>
<th>TYPE OF IMPACT</th>
<th>TYPE OF REGION OR ARCHETYPICAL SITUATION THAT COULD BE CONSIDERED</th>
</tr>
</thead>
<tbody>
<tr>
<td>Human toxicity (carcinogens + non-carcinogens)</td>
<td>Local</td>
<td>Air emission: High vs. average vs. low population density (for pollutants dominated by the inhalation pathway); Intensive vs. extensive vs. non-agricultural region (for pollutants dominated by the food pathway); Off-shore Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river Soil emission: Agricultural vs. non-agricultural soil</td>
</tr>
<tr>
<td>Respiratory effects</td>
<td>Local</td>
<td>Air emission: High vs. average vs. low population density; Off-shore caused by inorganics</td>
</tr>
<tr>
<td>Ionizing radiation</td>
<td>Local</td>
<td>Air emission: High vs. average vs. low population density (for pollutants dominated by the inhalation pathway); Intensive vs. extensive vs. non-agricultural region (for pollutants dominated by the food pathway); Off-shore Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river Soil emission: Agricultural vs. non-agricultural soil</td>
</tr>
</tbody>
</table>
### Table 1. (continuation)

<table>
<thead>
<tr>
<th>IMPACT CATEGORY</th>
<th>TYPE OF IMPACT</th>
<th>TYPE OF REGION OR ARCHETYPICAL SITUATION THAT COULD BE CONSIDERED</th>
</tr>
</thead>
<tbody>
<tr>
<td>O$_3$ layer depletion</td>
<td>Global</td>
<td>-</td>
</tr>
<tr>
<td>Photochemical oxidation</td>
<td>Local</td>
<td>Country; High vs. average vs. low population density</td>
</tr>
<tr>
<td>Aquatic ecotoxicity</td>
<td>Local</td>
<td>Air emission: Country; Off-shore Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river Soil emission: Type of watershed</td>
</tr>
<tr>
<td>Terrestrial ecotoxicity</td>
<td>Local</td>
<td>Air emission: Off-shore Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river Soil emission: Agricultural vs. non-agricultural soil</td>
</tr>
<tr>
<td>Terrestrial acidification/nutrification</td>
<td>Regional</td>
<td>Air emission: Country/continent Soil emission: Country/continent; Type of soil</td>
</tr>
<tr>
<td>Aquatic acidification</td>
<td>Regional</td>
<td>Air emission: Country/continent Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river; Type of lake</td>
</tr>
<tr>
<td>Aquatic eutrophication</td>
<td>Regional</td>
<td>Air emission: Country/continent; Water emission: Upstream vs. downstream of a lake; Ocean vs. lake vs. river; Type of lake</td>
</tr>
<tr>
<td>Land occupation</td>
<td>Local</td>
<td>Type of land; Country; Type of ecosystems</td>
</tr>
<tr>
<td>Biodiversity loss</td>
<td>Local</td>
<td>Type of land; Country; Type of ecosystems</td>
</tr>
<tr>
<td>Global warming</td>
<td>Global</td>
<td>-</td>
</tr>
<tr>
<td>Fossil energy</td>
<td>Global</td>
<td>Country (especially related to social issues)</td>
</tr>
<tr>
<td>Mineral extraction</td>
<td>Global</td>
<td>Country (especially related to social issues)</td>
</tr>
<tr>
<td>Noise</td>
<td>Local</td>
<td>High vs. average vs. low population density</td>
</tr>
<tr>
<td>Accidents</td>
<td>Regional</td>
<td>Country / continent</td>
</tr>
</tbody>
</table>

To capture the information inherent in spatial variability, *geographically differentiated* or *situation dependent* approach can be used (Potting & Hauschild 2004). Situation dependent refers to cases where spatial differences can be classified into a few archetypical situations e.g. agricultural or urban area, high vs. low population density area. It can better adapted to model local impact categories. Geographically differentiated means the use of regions in which the characteristics are uniform (continents, countries, or regions), and is used for regional impact categories.
1.3 REGIONALIZATION IN ECOTOXICITY ASSESSMENT OF METALS

In order to evaluate toxicity of different substances, the first LCIA models represented the recipient ecosphere as a generic medium with average properties. It has simplified the modelling of fate and exposure of the substance, so the characterisation factors (comparative toxicity potentials - CTPs) were much more dependent on the intrinsic toxic characteristics than where and how the substances are released (Heijungs 2012). However the recognition of importance of other site dependent factors such as bioavailability, persistence and so on resulted in the implementation of multi-media models for fate and exposure of chemicals, which models were widely used in human and environmental risk assessment (HERA) before (Heijungs 2012). Since the fate of chemicals depends on a huge number of parameters, such as soil properties, climatic conditions etc. these multi-media models has become more and more complex and spatially explicit. A current trend is the use of location specific conditions for assessing the exposure to metals (Mutel et al. 2011; Heijungs 2012). However, the optimal level of regionalization is always a debate, most recently Heijungs (2013) has criticised the recent trends of so called “hyperregionalization”, which term is interpreted as the inclusion of high level of spatial detail in LCIA resulting weaker applicability in LCA practice.

Because no scientific model can represent the reality perfectly, to reach the optimum model performance it is necessary to find equilibrium between the level of detail against the demand for the always uncertain input data (Sala et al.). The often missing availability of spatially resolved emission data and the computing costs of such models also need to be considered.

Coal-burning industrial processes generate a high amount of aerosols and the environmental effect of metals bound to such particles in neighbouring soils is very much depending on the soil properties. The ecological toxicity of metals in soil is determined by the bioaccessibility and bio-availability of a given metal for the living organisms and can vary several orders of magnitudes in different soil types. Since soil parameters can vary over geographic space very significantly, it is necessary to further develop spatially resolved models in order to reduce the uncertainty of results derived from the multimedia models without spatial resolution.
1.3.1 Deposition of airborne copper from coal-fired power plants

Despite of the last couple of decade's effort to improve air quality in European countries by applying stricter regulations and standards for filtering large chimneys, the coal-fired power plants still emit a large quantity of particulate matter (PM) every year. The size of the particles are relatively small (between 0.1 and 50 μm). When this particles or aerosol (with the copper bounded to it) leave the stack, sooner or later it deposit somewhere else depending on many factors. Generally the motion of the particle rely on the balance of different forces that are acting upon it. E.g. if the particle is bigger, heavier, the gravitation force will be higher than the others such as drag force, so it will fall down sooner, and closer to the emission source. However small particles (diameter less then 1 μm) can travel several thousands of kilometers before landing, because in that case the mixing effect of air turbulence is so great that the particles can easily ascend to even higher than the original stack height, spending several days or weeks in the atmosphere.

Meteorological conditions such as wind speed or atmospheric stability also can influence the turbulence and the drag force of the wind and precipitation can dramatically speed up deposition, because it washes out the aerosols from air. Emission stack parameters such as height, stack tip temperature, pressure etc. are also important factors controlling the shape of the plume that contains the contaminants. Even the receiving surfaces count as well, because some certain surfaces are more rough than others being able to adsorb aerosols more easily.

Atmospheric transport and deposition modeling are widely used to calculate dispersion and deposition of emitted substances using mathematical equations. From the beginning of the 20th century, there has been some investigations to numerically describe the phenomenon, however the rising interest from air pollution control regulations dating back in the late 1960's gave the real push to the spreading of such approaches. A number of method has been developed (e.g. Gaussian, Lagrangian, Eulerian, Box-type etc.) providing analytical and/or empirical solutions to the problem.
1.4 OBJECTIVES AND END GOAL

The aim of this thesis is to evaluate spatial variability in the CF of Cu when size of deposition areas, differences in deposition load as affected by distance from the source, and sensitivities of receiving soils, are taken into account. Cu emitted to air from 6 major power plants located in Hungary was chosen as a model metal due to the availability of models for calculating soil-specific CTP. The work consists of:

- the creation of rasterized data by applying the regression model introduced by Owsianiak et al. (2013) to obtain soil-specific comparative toxicity potentials (CTPs) for each raster cell, based on a soil properties (available from a global soil dataset);

- Developing weighting schemes to calculate source-specific CTPs that takes into account differences in deposition patterns, depending on the emission locations and atmospheric conditions. For that purpose, comparing and evaluating existing atmospheric dispersion models or developing one if necessary, and after harmonizing the model results linking the calculated deposition fields to the CTP grid obtained in the previous step.
• For case studies, all six coal-fired power plants in Hungary have been selected, having greater capacity than 50 MW, with the purpose of calculating country-specific CTPs. It means that the average terrestrial ecotoxicity of one unit emission of particle-bound copper emitted by Hungarian coal-fired electricity production in soils can be assessed based on the results of this work.
CHAPTER 2: METHODS

2.1 OVERVIEW

In this chapter the calculation and modeling techniques, case study locations and the data sources are described. First a very brief description of ecotoxicity characterisation is given, then the field study locations, atmospheric deposition models, and the calculation of comparative toxicity potentials follows.

The toxicity of e.g. 1 kg copper to soil can be very different if the receiving soil is sensitive to it (e.g. has lower pH) or just the opposite, has higher buffer capacity, higher pH etc. This variability is need to addressed somehow in the models. Of course very often there are no or limited information is available about the exact emission source, or the data has high uncertainty, so the model has weak applicability in everyday LCA practice (Heijungs 2013). In this thesis a solution is mentioned for the problem by averaging CTPs of soils for each emission source (in this case coal-fired power plants) with weighting with the deposition field obtained from atmospheric dispersion models.

Regarding the softwares used, the original goal was to reduce the number of applications to the minimum in order to keep the computation as simple as possible, however at some point in the data processing, additional work with other different applications could not be avoided. Thus we used Environmental Systems Research Institute (ESRI) ArcGIS (version 10.1) geographic information system (GIS) and geodatabase management application to make the Harmonized World Soil Database (HWSD) data available for calculations in Mathworks MATLAB, and also, in the final phase, to create the supporting thematic maps. Microsoft Access and Excel has been also needed to export soil properties from HWSD as described later. Althoug a couple of softwares has been used, the vast majority of the modeling work has been done using MATLAB (version 2013a) numerical computing environment with Mapping Toolbox and Image Processing Toolbox.

The only exception was the running of Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) atmospheric dispersion model, since it has its own software...
framework freely accessible from the developer, Air Resources Laboratory (ARL) website.\(^1\)

Figure 6 shows the general structure of the methods and data sources applied. First, soil property raster datasets has been created from Harmonized World Soil Database (HWSD) using the appropriate MS Access queries and connecting the results to the HWSD raster. Those rasters then used for calculating the soil-specific CTPs. In parallel deposition field are obtained by running different atmospheric dispersion models on the same grid with the same geographic location as the HWSD has. In this way linking or overlaying the two raster - in order to weight the CTPs - has made simple.

In this way, the more metal deposits to a soil, the higher the weight of it in the average resulting in one “plant-specific” average CTP per each of the facilities. The plant-specific CTP can be seen as the comparative toxicity potential of one unit of emitted particle-bound copper from the the given power plant in soil. If all plants has its own CTP value in a country, and the emitted mass is known for each of the plants, then a ‘country-specific’ average CTP can be calculated.

\(^1\) [http://ready.arl.noaa.gov/HYSPLIT.php](http://ready.arl.noaa.gov/HYSPLIT.php), access date: 09.04.2013
2.2 COMPARATIVE TOXITY POTENTIALS OF SOILS AND SOIL DATA

2.2.1 Calculation of comparative toxicity potentials

Calculation of CTPs of metals in soils after unit emission to air is entirely based on the method developed by Owsianiak et al. (2013). They presented it for calculation of CTP's of copper and nickel, that is why the modeling of copper emission has been chosen in this thesis. A short description is provided here, without going into much details.

In order to estimate which parameter (pH, clay content, organic carbon content etc.) controls the CTP mostly, first they calculated the CTP's for a set of 760 soil units. The widely accepted USEtox has been used to calculate fate factors, and terrestrial biotic ligand model (TBLM) for effect factor, while empirical regression models employed to predict bio-availability and accessibility factors.

Knowing the CTPs for the soils, a multiple linear regression (MLR) model has been used to analyse the importance of each soil property variables. Some parameters showed strong correlation with others, so those could be excluded. They found that the following equation can be used to calculate CTPs directly from soil parameters:

\[
\log_{10}(CTP) = a + b \cdot pH + c \cdot \log_{10}(\text{ORGC}) + d \cdot \log_{10}([\text{Mg}^{2+}]) + e \cdot \log_{10}(\text{CLAY})
\] (3)

where ORGC (%) is the organic carbon content; [Mg\(^{2+}\)] (mol/L) is magnesium concentration in soil pore water; and CLAY (%) is clay content, and \(a, b, c, d\) and \(e\) are empirical coefficients.

The best-fit parameters for the CTP equation can be seen in Table 2.

2. Table: Linear regression coefficients for calculating \(\log_{10}\) CTP. From Owsianiak et al. (2013). 'x' sign means the parameter is not used.

<table>
<thead>
<tr>
<th>a (intercept)</th>
<th>b (pH)</th>
<th>c ((\log_{10}(\text{ORGC})))</th>
<th>d ((\log_{10}([\text{Mg}^{2+}])))</th>
<th>e ((\log_{10}(\text{CLAY})))</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.327</td>
<td>x</td>
<td>-1.003</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>5.653</td>
<td>-0.408</td>
<td>-1.150</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>6.006</td>
<td>-0.426</td>
<td>-1.166</td>
<td>0.072</td>
<td>x</td>
</tr>
<tr>
<td>6.074</td>
<td>-0.427</td>
<td>-1.156</td>
<td>0.078</td>
<td>-0.035</td>
</tr>
</tbody>
</table>
Note that the more parameters are taken into account, the more accurate the estimate is.

Important limitations of the method presented above is that it can be applied for noncalcerous soils with pH value between 3.5-9, organic carbon below 38%, ionic strength of soil pore water below 0.5 M). In this thesis, soil units with values falling outside the mentioned limits were not excluded, but the effect of this assumption later will be discussed in Chapter 4.

2.2.2 Choosing soil database

To ensure the global coverage of applicability of our proposed method (which is a prerequisite in LCA) an easily accessible and global soil database has been needed. The best suitable for this purpose up to date is clearly the Harmonized World Soil Database (HWSD), which merged the publicly available data sources (SOTER, ESD, Soil Map of China, WISE) into one - harmonized - database. It is developed by a joint project consisting of the Food and Agriculture Organization of the United Nations (FAO), the International Institute for Applied Systems Analysis (IIASA), the International Soil Reference and Information Centre (ISRIC), Institute of Soil Science – Chinese Academy of Sciences (ISSCAS) and the Joint Research Centre of the European Commission (JRC).

HWSD is a raster data source with a resolution of 30 arc-second or approximately 1x1 km. One raster cell or pixel point can have up to 10 soil types (soil mapping units) with their approximate area percentages. In all there are around 16000 different soil mapping units in the HWSD. All soil mapping units has its uniqe set of phisico-chemical property values which consist of 17 separate variable - including silt, sand and clay fraction, bulk density, pH, organic carbon content etc. for top soil and the same parameters for sub soil as well.

To calculate CTPs, coefficients from the second row of the Table 2 has been used in Equation 3, because no [Mg$^{2+}$] concentration in soil pore water data is given in HWSD. The following pages shows the map of Europe as an example with the raster-cell averaged pH and ORGC content map. Relative occurrence of soil mapping units

2.2.2.1 Exporting data from HWSD

After downloading the HWSD data from the International Institute for Applied Systems
Analysis's (IIASA) website\(^2\), and unzipping, it is possible to directly load the included raster file (hwsd.bil) into ArcGIS, since it uses the ESRI's BIL format. We followed the technique described in the HWSD manual Annex 3 ('Use of the HWSD in GIS software').

Map 1. Topsoil pH in Europe. Source: HWSD 2012. Scale 1: 30 000 000.

Legend:
- NA
- 3.01 - 4
- 4.01 - 5
- 5.01 - 6
- 6.01 - 7
- 7.01 - 8
- 8.01 <

Coordinate system: GCS WGS84.
Map 2. Topsoil organic carbon content in Europe [m/m%]. Source: HWSD 2012.
Scale 1: 30 000 000

Legend
- NA
- < 0.5
- 0.51 - 1
- 1.01 - 2
- 2.01 - 5
- 5.01 - 10
- 10.01 - 20
- 20.01 <

Coordinate system: GCS WGS84
2.3 CASE STUDY LOCATIONS

As case study locations, coal-fired power plants has been selected, because they are well known large emitters of particle-bound copper (Smolka-Danielowska 2006), since the fuel (usually brown coal or lignite) usually contains copper as a trace metal. Compared to other sources of particulates such as transport they have the advantage of having exact location, which make the air deposition modeling much easier. In 2010 Hungary there were six large-scale (capacity over 50 MW) coal-fired power plant, according to (MAVIR 2011) (see Map 4 and Table 4). One of them was out of order in 2010, so it has been excluded from the analysis.

Table 3: Basic data of large-scale coal-fired power plants in 2010 in Hungary listed by annual electricity production. * excluded plants source (MAVIR 2011)

<table>
<thead>
<tr>
<th>Power plant</th>
<th>Capacity (MW)</th>
<th>Electricity production (TJ)</th>
<th>Heat production (TJ)</th>
<th>Brown coal usage (TJ)</th>
<th>Biomass usage (TJ)</th>
<th>Constr. date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mátrai</td>
<td>950</td>
<td>22636</td>
<td>231</td>
<td>55333</td>
<td>7857</td>
<td>1969-72</td>
</tr>
<tr>
<td>Oroszlány</td>
<td>240</td>
<td>3143</td>
<td>374</td>
<td>6968</td>
<td>3888</td>
<td>1961-62</td>
</tr>
<tr>
<td>Tiszapalkonya</td>
<td>200</td>
<td>1231</td>
<td>820</td>
<td>5291</td>
<td>0</td>
<td>1955-62</td>
</tr>
<tr>
<td>Borsod</td>
<td>136.9</td>
<td>932</td>
<td>0</td>
<td>23</td>
<td>4381</td>
<td>1951-57</td>
</tr>
<tr>
<td>Ajka</td>
<td>101.6</td>
<td>626</td>
<td>2523</td>
<td>2690</td>
<td>2874</td>
<td>1957-57</td>
</tr>
<tr>
<td>Tatabánya*</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1950-54</td>
</tr>
</tbody>
</table>

Map 3. Location and capacity of the Hungarian coal-fired power plants, that are included in the study. The size of circle is proportional to the combined electricity and heat production in 2010.
2.4 ATMOSPHERIC DISPERSION AND DEPOSITION

Three different approaches has been selected and compared in this thesis:

- a simple exponential decline deposition function, developed by De Caritat et al (1997);
- the author's modification of a Gaussian type steady-state plume, which is somewhere between the two abovementioned models from computational viewpoint (much simpler than the third one, but more complex than the 'De Caritat' model),
- and a very sophisticated atmospheric model, Hybrid Single Particle Lagrangian Integrated Trajectory (HySPLIT,) which is able to predict even hourly concentrations.

Each of the models were applied in an 500 km circle around the emission sources, because within this distance the deposition of the vast majority of the particles are expected.

### 2.4.1 Model 1: 'De Caritat'

De Caritat et al. (1997) presented a relatively simple model for airborne trace metals emitted from smelters. Consequently, in order to apply this approach for the case of power plants, the particles from smelters assumed to behave similarly to those from from power plants.

Based on field measurements of heavy metal concentration in mosses around point sources in the Kola Peninsula, Russia, they found that the deposition of particles occurring mostly within 200 km distance, following an exponential decline function. In the closest vicinity of the stack there is a 'shadow' zone where less metal falls. They created a function for the annual deposition profile along one direction from the source then parameterised it using field data. Then the loading function (kg deposited mass km$^{-2}$ year$^{-1}$) has obtained by integrating the deposition-distance function around the emission...
source, so the result is a relatively simple, circular shape deposition field, with empirical variables. $D_1$ is the deposition (kg km$^{-2}$) at 1 km distance in from the source, while $\kappa$ is a dispersion coefficient which determines the steepness of the curve and depends on the industry and the specific contaminant. The loading function is a linearly increases from the source to $D'$:

$$D(x) = \left( \frac{D' - D_0}{x'} \right) x + D_0 \quad [\text{kg km}^{-2}]$$

where $D_0$ is the deposition at $x = 0$ (kg km$^{-2}$), $x'$ critical distance where maximum loading occurs (km), and $D'$ is the deposition at $x'$ (kg km$^{-2}$).

In the outer domain from $D'$ to the remote areas the function decreases as a power law of the distance:

$$D(x) = D_1 x^{\kappa} \quad [\text{kg km}^{-2}]$$

where $D_1$ deposition at 1 km distance from the source (kg km$^{-2}$), $\kappa$ coefficient (-).

![Diagram of the deposition-distance relationship](image)

**Figure 7. The scheme of the deposition-distance relationship (from De Caritat et al. 1997)**
Theoretically $D_0$, $x'$ and other parameters depends on the industrial activity type, metal contaminant etc. However I could not be able to find similar estimates for my cases, so their originally published values has been used in this thesis (see Table 4). Their parameters has been validated for their special case by field measurement fitting, so it could be a source of error.

Table 4: Best-fit parameters of deposition-distance relationship for copper from De Caritat et al. 1997

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$\kappa [-]$</th>
<th>$D_1 [\text{kg km}^{-2}]$</th>
<th>$x' [\text{km}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>-2.0031</td>
<td>14220.67</td>
<td>0.1-0.3</td>
</tr>
</tbody>
</table>

2.4.2 Model 3: Gaussian plume

First (Sutton 1947) derived an air pollutant plume dispersion model from the well-known convection-diffusion equation with the assumption of Gaussian distribution for the vertical and crosswind dispersion of the plume. From the beginning, Gaussian plume type models have been the most widespread due to their computational simplicity, yet well agreement with the experimental data (Hanna et al. 1982).

Normally these models has been applied for gases and aerosols, in which cases gravity does not influence greatly the motion of the pollutants. Ermak (Ermak 1977) provided a derivation which takes into account gravitational settling and dry deposition of particulate matter also.

2.4.2.1 Pollutant concentration in the air

According to the abovementioned work (Ermak 1977) the concentration formula from a point source located in $(0,0,h)$:
\[ C(x, y, z) = \frac{Q}{2\pi U \sigma_y \sigma_z} \exp\left(\frac{-y^2}{2\sigma_y^2}\right) \exp\left(\frac{-V (z-h)}{2K_z} \right) \frac{V^2 \sigma_z^2}{8K_z^2} \cdots \]
\[ \times \left[ \exp\left(\frac{-(z-h)^2}{2\sigma_z^2}\right) \right. \left. \exp\left(\frac{-(z+h)^2}{2\sigma_z^2}\right) \right] ^{\sqrt{2\pi}} W_0 \sigma_z \sigma_z \exp\left(\frac{W_0 \sigma_z}{\sqrt{2}K_z} + \frac{(z+h)}{\sqrt{2}\sigma_z}\right) \]
\[ W_0 = V - \frac{1}{2} W \]

where \( C \) steady state downwind pollutant concentration at \((x, y, z)\) (kg m\(^{-3}\)), \( Q \) constant emission rate or source strength (kg s\(^{-1}\)), \( U \) effective wind speed (m s\(^{-1}\)), \( \sigma_y, \sigma_z \), horizontal and vertical diffusion parameters (-), \( K_z \), eddy diffusion coefficient (m\(^2\) s\(^{-1}\)), \( V \) deposition velocity (m s\(^{-1}\)), \( W \) constant settling velocity (m s\(^{-1}\)), \( h \) stack height (m), \( \text{erfc}(x) \) complementary error function.

**Inherent assumptions in the model:**

- Steady state plume, meaning that the emission rate \( Q \) is constant and continuous from a single point source, and wind speed \( U \) is constant and the wind blows in the \( x \)-direction:
  \[ \delta Q/\delta t = 0; \delta U/\delta t = 0; U_y, U_z = 0; \]
  \[ \text{(5)} \]

- Eddy diffusivities (\( K_x, K_y, K_z \)) are the functions only the downwind distance:
  \[ K(x,y,z) = K(x), \]
  \[ \text{(6)} \]

- Constant wind speed with height:
  \[ \delta U/\delta z = 0; \]
  \[ \text{(7)} \]

**Other assumptions:**

- No plume 'reflection' off of ground or the top of the mixed layer;
• No stack induced plume downwash (effect of lower pressure near the stack);

• No buoyant plume rise.

2.4.2.2 Determining parameters for calculating the pollutant concentration

Diffusion parameters

Diffusion parameters $\sigma_y$ and $\sigma_z$ are the standard deviations of the distribution of the concentration in the $y$ and $z$ directions, respectively (Hanna et al. 1982). There are many formulae and semi-empirical expressions available for determining $\sigma_y$ and $\sigma_z$ under different conditions of atmospheric stability. In this work the so-called Pasquill-Guifford (P-G) scheme has been adopted, which has been used elsewhere (Winges, 1991) for this type of modeling. In this approach the atmospheric stability has been classified into six category, depending on the solar radiation and average wind speed and surface type (rural or urban). Each class has a label from A to F, where A is the least stable (greatest turbulent mixing).

Following the method used in the US Environmental Protection Agency (EPA) Industrial Source Complex (ISC3) model (EPA 1995), the diffusion parameters can be determined using equations [2,3]:

$$\sigma_y = 465.11628 \tan[0.017453293(a_y - b_y \ln(x))] [m] \quad (8)$$

where $x$ downwind distance, $a_y$ and $b_y$ parameters depending on atmospheric stability class, and

$$\sigma_z = a_z x^{b_z} [m] \quad (9)$$

$a_z$, $b_z$ parameters depending on atmospheric stability class, and downwind distance. The exact empirically obtained values of $a_y$, $b_y$, $a_z$, $b_z$ can be found in the ISC3's User's Guide (EPA 1995). See Table 1 and 2 in Appendix for estimates of parameters.

To simplify the model and to reduce data requirements an assumption of constant stability class 'D' and rural terrain has been made, because it seemed reasonable in average meteorological conditions. However in the MatLab script, the possibility of changing the P-
G class has been implemented to later assess the sensitivity towards input parameters.

2.4.2.3 Eddy diffusivity

Eddy diffusion is the process of air pollutant mixing due to eddy motion. To calculate the coefficient $K$, which can parametrise this phenomenon a similar method has been used as in EPA’s Fugitive Dust Model (FDM) (Winges 1991) and (Lushi & Stockie 2010):

$$\sigma_z^2(x) = \frac{2}{U} \int_0^x K_z(x') dx' \quad [\text{m}^2\text{s}^{-1}] \quad (10)$$

since K assumed constant, then

$$K_z = \sigma_z^2 U / 2x \quad (11)$$

2.4.2.4 Settling velocity

Settling velocity is approximated by Stokes’ Drag Formula for spherical objects:

$$W = \frac{\rho g d^2}{18\mu} \quad (12)$$

where $W$ settling velocity (m s$^{-1}$), $\rho$ particle density (kg m$^{-3}$), $g$ gravitational acceleration (ms$^{-2}$), $d$ particle diameter (m), $\mu$ dynamic viscosity of air (Pa s).

For particles smaller than 15 $\mu$m is is necessary to introduce the Cunningham slip factor:

$$W_C = \frac{W}{f_{\text{shape}}}(1 + 2 \frac{\lambda}{d}(1.26 + 0.4 e^{-0.55 \frac{d}{\lambda}})) \quad (13)$$

where $W_C$ is the corrected settling velocity (m s$^{-1}$), $d$ particle diameter (m), $f_{\text{shape}}$ shape factor - largest diameter per smallest diameter (-), and $\lambda$ is free mean path of air particles.
2.4.2.5 Deposition on the ground

2.4.2.5.1 Deposition flux

Boundary condition for dry deposition on the ground following (Lushi & Stockie 2010; Winges 1991):

\[
[K \frac{\partial C}{\partial z} + WC]_{z=0} = VC \big|_{z=0} = F_{\text{dry}} \quad \text{[kg m}^{-2} \text{s}^{-1}] \tag{14}
\]

where \( V \) is deposition velocity. Pollutant deposition rate onto the surface is proportional to the local air concentration. In a given location \( F_{\text{dry}} \) is simply the product of the local concentration \( C \) and the deposition velocity \( V \):

\[
F_{\text{dry}}(x,y) = VC_{x,y} \tag{15}
\]

2.4.2.5.2 The deposition per unit time or deposition rate

Deposition rate in a receptor located at position \((r_i)\) having a cross-sectional area \( A \) (assumed small enough) is approximated by:

\[
I_{\text{dry}} = \int_A - AF_{\text{dry}}(x, y, 0) \, dx \approx -AF_{\text{dry}}(r_i) = AV C_{x,y} \quad \text{[kg s}^{-1}] \tag{16}
\]

2.4.2.5.3 Wet deposition

The calculation of wet deposition is entirely based on (Hanna et al. 1982). Wet deposition flux (including both rainout and washout) \( F_{\text{wet}} \) is found by using the scavenging ratio:

\[
F_{\text{wet}} = CWrJ \quad \text{[kg m}^{-2} \text{s}^{-1}] \tag{17}
\]

where \( Wr \) scavenging ratio [-]. The physical meaning of \( Wr \) is the ratio between the concentration of particles in precipitation and concentration in air, \( J \) precipitation rate (m s\(^{-1}\)). Empirical values of \( Wr \) values are summarized in (McMahon & Denison 1979).

Wet deposition per unit time similar to eq. (9):
2.4.2.6 Unit time and treatment of varying wind

Eq. (1) are derived for steady state conditions (the wind is constant over time interval $\Delta t$). However the wind speed $U$ and direction $\theta$ changes all over the year, which has a significant influence on the deposition field. Since monthly mean wind speed and direction distribution data is easily accessible (e.g. [http://www.windfinder.com](http://www.windfinder.com)), we divide time into equally-spaced interval $\Delta t = T/N$ assuming $U(t)$ and is constant on each interval. The wind direction distribution is measured in 16 direction, but for a better resolution, the distribution is interpolated for intermediate angle intervals.

The method described by (Lushi & Stockie 2010) and has been adapted to our goal with a slight modification. The deposition (either dry or wet) for one direction with $N$ measurements of average wind speed:

$$D_1(x, y) = \sum_{i}^{N} \Delta t_n \ I_N(C(x, y, U_N, ...)) \ [\text{kg}] \quad (19)$$

where $\Delta t_n$ time interval [s], $N$ number of temporal wind speed averages. Wet deposition occurs over $t_{\text{wet}}$ [h] and it is the product of annual rainfall $P$ [mm] and an assumed average rainfall rate $J$ [mm h$^{-1}$]:

$$t_{\text{wet}} = P \ J \ = \ (356 \cdot 24) - t_{\text{dry}} \ [\text{min}] \quad (20)$$

where $t_{\text{dry}}$ the time interval when dry deposition occurs (min). After calculating for one direction, the plume is rotated by the windrose directions and weighting with its distribution:

$$D(x, y) = \sum_{M}^{M} \Theta_M \ D_M((x \cos 2\theta_M - y \sin 2\theta_M), (x \sin 2\theta_M - y \cos 2\theta_M)) \ [\text{kg}]$$

$$\sum_{M}^{M} \Theta_M = 1 \quad (21)$$

where $\Theta_M$ the relative frequency of $M^{\text{th}}$ wind direction angle [-], $\theta_M$ is the $M^{\text{th}}$ wind direction angle [rad]. Thus, the treatment of time variation can be seen as solving a sequence of steady-state plume problem, and the final deposition field is obtained by summing the
results over each time interval.

2.4.2.7 Total deposition field for Cu

The total deposition of copper has been obtained by multiplying the deposited mass of PM with the copper concentration in it:

\[ D_{Cu}(x,y) = D(x,y) \cdot c_{Cu} \text{ [kg]} \]  \hspace{1cm} (22)

where \( c_{Cu} \) is the concentration of the Cu in the emitted particulates [kg/kg]. Since no measurement were available from the emission sources, then 20 ppm has been assumed based on the litterature (Halliburton et al. 2006; Smolka-Danielowska 2006). That means that the deposition field for 1 kg of PM or for 1 kg of particle-bound Cu does not differ from each other, since the calculation provided deposition field for a unit emission.

2.4.2.8 Overview of the model

\[ \begin{array}{c}
\text{Grav. accel., } g \\
\text{Particle density, } \rho \\
\text{Viscosity of air, } \mu \\
\text{Particle diameter, } d \\
\text{Downwind dist., } x \\
\text{Atm. stability class} \\
\text{Pasquill-G. coeff.} \\
\hline
\text{Stack height, } h \\
\text{Settling velocity, } W \\
\text{Emission rate, } Q \\
\text{Source location} \\
\text{Dep. velocity, } V \\
\text{Diff. parameters, } \sigma \\
\text{Eddy diffusivity, } K \\
\text{Wind speed, } U \\
\text{Scaveng. ratio, } Wr \\
\text{Unit time, } \Delta t \\
\text{Windrose, } \Theta \\
\text{Grid size, } A \\
\text{Dep. field (dry)} \\
\text{Dep. field} \\
\text{Dep. field (wet)} \\
\hline
\text{Dep. flux (wet), } F_{\text{wet}} \\
\text{Annual rainfall, } P \\
\text{Rain intensity, } J
\end{array} \]

Figure 8. Structure of the Gaussian-type atmospheric deposition model. Calculated variables are in grey, input parameters are in white box.
Site-specific parameters can be found in Table 8. Location has been obtained from literature. I downloaded wind data, and annual rainfall from freely available internet database. Due to the lack of data, other parameters has been assumed the same in every location (see Table 5 for site-generic parameters).

Table 5: Site-generic input parameters for the Gaussian-type model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Source / comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deposition velocity</td>
<td>V</td>
<td>$6.5 \times 10^{-4}$</td>
<td>m s$^{-1}$</td>
<td>(McMahon &amp; Denison 1979; T. M. Bachmann 2006)</td>
</tr>
<tr>
<td>Atmospheric stability class</td>
<td>-</td>
<td>D</td>
<td>-</td>
<td>assumed</td>
</tr>
<tr>
<td>Pasquill-Gifford coefficients</td>
<td>$a_y$, $b_y$, $a_z$, $b_z$</td>
<td>[see in text]</td>
<td>-</td>
<td>(EPA 1995)</td>
</tr>
<tr>
<td>Gravitational acceleration</td>
<td>g</td>
<td>9.81</td>
<td>m s$^{-2}$</td>
<td>-</td>
</tr>
<tr>
<td>Particle density</td>
<td>$\rho$</td>
<td>1000</td>
<td>kg m$^{-3}$</td>
<td>assumed, similarly to (Yun et al. 2002)</td>
</tr>
<tr>
<td>Viscosity of air</td>
<td>$\mu$</td>
<td>$1.8 \times 10^{-5}$</td>
<td>kg m$^{-1}$ s$^{-1}$</td>
<td>-</td>
</tr>
<tr>
<td>Particle size</td>
<td>d</td>
<td>$2 \times 10^{-7}$ - $4 \times 10^{-5}$</td>
<td>m</td>
<td>(T. M. Bachmann 2006) first set</td>
</tr>
<tr>
<td>Stack height</td>
<td>h</td>
<td>60</td>
<td>m</td>
<td>assumed</td>
</tr>
<tr>
<td>Scavenging ratio</td>
<td>Wr</td>
<td>$1 \times 10^{-6}$</td>
<td>-</td>
<td>(Bachmann 2006)</td>
</tr>
<tr>
<td>Rain intensity</td>
<td>J</td>
<td>5</td>
<td>mm h$^{-1}$</td>
<td>(Hanna, Briggs, and Hosker 1982)</td>
</tr>
</tbody>
</table>

Table 6: Site-specific input parameters for the Gaussian-type model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value range</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wind direction distribution</td>
<td>$\Theta$</td>
<td>-</td>
<td>%</td>
</tr>
<tr>
<td>Average wind speed at source</td>
<td>$U$</td>
<td>3-3.3</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>Annual rainfall</td>
<td>$P$</td>
<td>912-1139</td>
<td>mm</td>
</tr>
</tbody>
</table>

In Table 7, all the calculated variables are shown with the equation where the variables turn up.
Table 7: Overview of calculated variables

<table>
<thead>
<tr>
<th>Calculated variable</th>
<th>Symbol</th>
<th>Unit</th>
<th>Equation(s)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu deposition field</td>
<td>$D_{\text{Cu}}(x,y)$</td>
<td>kg</td>
<td>19-22</td>
<td>(Lushi &amp; Stockie 2010)</td>
</tr>
<tr>
<td>Dry deposition rate</td>
<td>$I_{\text{dry}}(x,y)$</td>
<td>kg s$^{-1}$</td>
<td>14-16</td>
<td>(Lushi &amp; Stockie 2010)</td>
</tr>
<tr>
<td>Wet deposition rate</td>
<td>$I_{\text{wet}}(x,y)$</td>
<td>kg s$^{-1}$</td>
<td>18</td>
<td>(Hanna et al. 1982)</td>
</tr>
<tr>
<td>Wet deposition flux</td>
<td>$F_{\text{wet}}(x,y)$</td>
<td>kg m$^{-2}$ s$^{-1}$</td>
<td>17</td>
<td>(Hanna et al. 1982)</td>
</tr>
<tr>
<td>PM concentration in air</td>
<td>$C(x,y)$</td>
<td>kg m$^{-3}$</td>
<td>4</td>
<td>(Ermak 1977)</td>
</tr>
<tr>
<td>Settling velocity</td>
<td>$W$</td>
<td>m s$^{-1}$</td>
<td>12,13</td>
<td>Stokes' Law for spherical objects</td>
</tr>
<tr>
<td>Eddy diffusivity</td>
<td>$K$</td>
<td>m$^{2}$ s$^{-1}$</td>
<td>6,10,11</td>
<td>(Winges 1991; Lushi &amp; Stockie 2010)</td>
</tr>
<tr>
<td>Diffusion parameters</td>
<td>$\sigma_y$, $\sigma_z$</td>
<td>m</td>
<td>8,9</td>
<td>(EPA 1995; Winges 1991)</td>
</tr>
</tbody>
</table>

2.4.3 Model 2: Hysplit

The HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) is an atmospheric dispersion and deposition simulation model with its own software package, developed by the United States National Oceanic and Atmospheric Administration (NOAA) (Draxler & Rolph 2013). Its usual simulation time frame is a couple of days, however it is also able to simulate over much longer periods of time (e.g. in (Chen & Stein 2013)). The HYSPLIT's built-in core computes single air parcel (particle or gas) trajectories over 3D space and time using hourly distributed complex meteorological datasets. The software is freely available through its website (http://ready.arl.noaa.gov/HYSPLIT.php). It has the capability of downloading preformatted meteorology data files for the simulations as well.

Since the objective is to obtain annual deposition field in this work, the NOAA National Centers For Environmental Prediction NCEP and the National Center for Atmospheric Research (NCAR/NCEP) joint reanalysis data set has been chosen as meteorological data source, because its 6-hourly and daily measurement frequency is well enough for the goal (instead of choosing other available higher resolution e.g. 1-hourly datasets). It uses
a 2.5 x 2.5 degree latitude / longitude raster grid, and contains a global analysis starting from 1948 using a single analysis system for the entire dataset, packaged together to monthly data files.

Beyond the weather input files, a number of other parameters can be set up in the HYSPLIT model. For the particle size distribution I followed (T. M. Bachmann 2006), who gave five different particle size groups with its subsequent mass shares, settling velocities and scavenging ratios. The concentration of copper is assumed the same regardless of particle size (Halliburton et al. 2006). The simulation timeframe has set to cover the year 2010.

Table 6 a summarizes the manually set up parameters below.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
<th>Source / comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle sizes (5 class)</td>
<td>$2 \times 10^{-7}$</td>
<td>m</td>
<td>(Bachmann 2006)</td>
</tr>
<tr>
<td>Deposition velocity</td>
<td>$6.5 \times 10^{-4}$</td>
<td>m s$^{-1}$</td>
<td>(McMahon and Denison 1979; Bachmann 2006)</td>
</tr>
<tr>
<td>Emission source location</td>
<td>m</td>
<td></td>
<td>not the same as geographical location</td>
</tr>
<tr>
<td>Particle density</td>
<td>1000</td>
<td>kg m$^{-3}$</td>
<td>assumed, similarly to (Yun, Yi, and Kim 2002)</td>
</tr>
<tr>
<td>Particle shape factor</td>
<td>1</td>
<td>-</td>
<td>assumed</td>
</tr>
<tr>
<td>Scavenging coefficients</td>
<td>$1 \times 10^{-6}$</td>
<td>s$^{-1}$</td>
<td>(Bachmann 2006)</td>
</tr>
<tr>
<td>Stack height</td>
<td>60</td>
<td>m</td>
<td>assumed</td>
</tr>
</tbody>
</table>

2.5 AREA-WEIGHTED AND COUNTRY-WEIGHTED CTPS

To calculate area-weighted CTPs the following formula has been used:

$$aCTP_n = \sum_{i=1}^{m} aWF_i CTP_i$$

(23)

where $aCTP_n$ is the area-weighted comparative toxicity potential emitted from the $n$th source (m$^3$/kg$_{total \ emitted \ to \ air \cdot \ day}$), $CTP_i$ is the comparative toxicity potential in soil of $i$th cell (m$^3$/kg$_{total \ emitted \ to \ air \cdot \ day}$), $aWF_i$ is the weighting factor for the $i$th cell (-).
Calculating weights seems to be an easy task, because one can say that as much of the copper deposits in a cell as high the weight is. However, several times the deposition model can not count for all of the emitted mass, because there are always some particles that can travel further away than the area investigated. Or it is possible that the emitted PM ends up in the ocean or a lake. That is why I decided to divide the calculated CTPs by the sum of the weights:

\[
aWF_i = \frac{A_i D_i}{\sum_{i=1}^{m} A_i D_i}
\]  

(24)

where \(A_i\) is the area of the \(i^{th}\) cell \([m^2]\), \(D_i\) is the deposition rate at the \(i^{th}\) cell \([kg \ m^{-2}]\), \(m\) number of raster cells. Introducing \(A_i\) is necessary because the HWSD grid cells are distributed equally by geographic degrees, so the cell area size gradually decreases as over distance from the equator. Then \(A_i \cdot D_i\) gives the ratio of mass unit that deposits within the raster cell.

Having the CTPs for all power plants in a given country, then

\[
cCTP = \sum_{n=1}^{k} WF_n aCTP_n
\]  

(25)

where \(cCTP\) country-weighted CTP \((m^3/kg_{total \ \text{emitted to air} \cdot \text{day}})\), and \(WF_n\) weighting factor of \(n^{th}\) power plant, \(k\) number of power plants in total. To obtain the weighting factors for each emission source, using annual power production seems to be a good approximation:

\[
WF_n = \frac{P_n EF_n}{\sum_{n=1}^{k} P_n EF_n}
\]  

(26)

where \(P_n\) annual combined heat and electricity power production of the \(n^{th}\) plant \((TJ)\), \(EF_n\) effect factor expressing the rate of copper emission per unit power generated \((kg \ Cu \ emitted / TJ \ power \ produced)\).

### 2.6 Sensitivity Analysis

In order to analyse the sensitivity towards different input parameters, I set up eight different scenarios, changing the variables one-by-one. For stack height, wind speed, and particle density, a realistic minimum and maximum value of the given variable have been
used, while for particle size distribution a different - coarser - distribution is assumed based on Bachmann (2006). For annual rainfall only a minimum value tested, because the given year - 2010 - was one of the most precipitation-abundant year in the meteorological measurement history in Hungary.
CHAPTER 3: RESULTS

3.1 SOIL SPECIFIC CTPS

Using the regression model (by Owsianiak et al. 2013) described in the previous chapter, the CTPs has been calculated for each cell. Map 4 shows the results for Europe. Comparing it to the previous maps (pH and organic carbon content) shows the expected effect of decreasing pH and organic carbon content on the increase of CTPs. However the two soil property follow an opposite general trend in the geographic space in Europe: pH tends to rise from north to south, while organic carbon content is gradually reducing, canceling out the general north-south directional differences, thus amplifying the small regional inequalities. The high variability of CTPs are apparent, often in the range of 3 orders of magnitude between close regions. Even in Hungary e.g. the easternmost corner has hundreds times higher CTPs than the soils in the neighbouring area.

3.2 DEPOSITION FIELDS OF CU ORIGINATING FROM POWER PLANTS

For all five locations, the deposition has been modeled using the three different atmospheric dispersion models. Map 5, 6 and 7 shows the resulting patterns for one location as an example (De Caritat, Gaussian and Hysplit, respectively), while Map 8 depicts all fields in one figure. Unit is mass relative to one emitted mass unit (MU/m²).

Map 5. Example map of airborne Cu annual deposition field using 'De Caritat' model, originating from Ajka power plant, 2010
Map 6. Example map of airborne Cu annual deposition field using Gaussian model originating from Ajka power plant, 2010

Map 7. Example map of airborne Cu annual deposition field using HYSPLIT model originating from Ajka power plant, 2010
Map 8. 500 km radius deposition fields of particle-bound Cu originating from different Hungarian power plants.
From left to right: De Caritat, Gaussian, Hysplit (atmospheric dispersion models)
From up to down: Ajka, Borsod, Mátrai, Oroszlány, Tiszapalkonya (power plants)
Unit: Mass unit per m²
Map 4. Soil-specific Comparative Toxicity Potentials in Europe (m³/kg (total emitted to air) · day).
Calculation based on Owsianiak et al 2013. Scale 1:30 000 000. Coordinate system: GCS WGS84
3.3 AREA-WEIGHTED CTPS

Applying the weighting formula given in equations 23-24, the emission source specific area weighted CTPs have been obtained across the 500 km distance from the emission source. As Figure 9 shows, this method reduced the original 2-3 orders of magnitude variability to around 0.5 after weighting.

![Calculated area-weighted CTPs for each location, with ranges of minimum and maximum values within 500 km distance from source at 95% variability interval. Average values of the results from the three different model are indicated by horizontal ticks. Red lines show the minimum and maximum values of soil specific CTPs from Owsianiak et al. (2013).](image)

Table 9. contains the calculated values and the percentage of emitted mass, that deposits within 500 km according to each model types.

<table>
<thead>
<tr>
<th>Power plant</th>
<th>Hysplit</th>
<th>Gaussian</th>
<th>De Caritat</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ajka</td>
<td>5.405e3 (71.52)</td>
<td>5.1334e3 (96.27)</td>
<td>4.5833e3 (60.24)</td>
</tr>
<tr>
<td>Borsod</td>
<td>3.8057e3 (69.95)</td>
<td>1.4218e3 (99.5)</td>
<td>2.3696e3 (58.69)</td>
</tr>
<tr>
<td>Mátrai</td>
<td>2.8729e3 (72.6)</td>
<td>1.9949e3 (99.8)</td>
<td>2.2572e3 (58.66)</td>
</tr>
<tr>
<td>Tiszai</td>
<td>3.3391e3 (70.93)</td>
<td>1.7921e3 (99.98)</td>
<td>2.3906e3 (58.66)</td>
</tr>
<tr>
<td>Vértesi</td>
<td>3.6572e3 (72.8)</td>
<td>2.2832e3 (94.44)</td>
<td>2.7588e3 (59.43)</td>
</tr>
</tbody>
</table>
3.4 COUNTRY WEIGHTED CTP

The calculation of plant-specific weights using eq. 26 is not possible in this case, because there are no plant specific emission factors available in the literature, so I assumed equal emission factors. Since the age of the plants are very similar, as well as the fuel burned, then it seems a reasonable assumption. Eq. 26 then reduces to:

\[ WF_n = \frac{P_n}{\sum_{n=1}^{k} P_n} \]

which means a simple weighting by the annual power generation.

Table 10: Country-weighted CTP of Cu emitted to air from Hungarian coal-fired power plants, based on different weighting schemes

<table>
<thead>
<tr>
<th>Model</th>
<th>Log10 (CTP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hysplit</td>
<td>3.51</td>
</tr>
<tr>
<td>Gaussian</td>
<td>3.36</td>
</tr>
<tr>
<td>De Caritat</td>
<td>3.41</td>
</tr>
</tbody>
</table>

It is also interesting, if we assume that all 1.5 % of the 60 kt emitted PM / year can be attributed to power plants (KSH 2010), and the PM emissions are proportional to the abovementioned annual production (in TJ), knowing the Cu concentration in the particulates (e.g. (Smolka-Danielowska 2006)) then one can approximate the total loading of Cu originating from Hungarian power-plants (see Map 9) using the deposition fields.

3.5 SENSITIVITY ANALYSIS

Surprisingly, none of the models are sensitive to emission stack height (Table 11). The reason might be found in the particle size distribution: very small particles (below 1um) do not settle close to the source and can be easily lifted up by the turbulence. the most precipitation-abundant year in the meteorological measurement history in Hungary.
Table 11: Area weighted CTPs in soil. In brackets the coverage of total emitted mass in %
*very small resulting area

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Gaussian</th>
<th>HYSPLIT</th>
</tr>
</thead>
<tbody>
<tr>
<td>original values</td>
<td>2.091e+03 (89.26)</td>
<td>2.3737e+03 (95.47)</td>
</tr>
<tr>
<td>particle size distribution scheme B</td>
<td>1.0669e3 (89.45)</td>
<td>2.5376e3 (80.16)</td>
</tr>
<tr>
<td>stack height = 30 m</td>
<td>2.3705e3 (95.74)</td>
<td>2.7221e3 (73.53)</td>
</tr>
<tr>
<td>stack height = 90 m</td>
<td>2.3822e3 (94.49)</td>
<td>2.9432e3 (70.23)</td>
</tr>
<tr>
<td>particle density = 500 kg/m3</td>
<td>2.3403e3 (99.78)</td>
<td>2.7300e3 (73.02)</td>
</tr>
<tr>
<td>particle density = 2000 kg/m3</td>
<td>2.3723e3 (99.6)</td>
<td>2.7300e3 (73.02)</td>
</tr>
<tr>
<td>precipitation = 400mm/a</td>
<td>2.3671e3 (55.56)</td>
<td>Not applicable</td>
</tr>
<tr>
<td>average wind speed = 2 m/s</td>
<td>1.8048e+03 (96)*</td>
<td>Not applicable</td>
</tr>
<tr>
<td>average wind speed = 5 m/s</td>
<td>2.3709e+03 (58.9)</td>
<td>Not applicable</td>
</tr>
</tbody>
</table>
Map 9. Approximate airborne Cu loading originating from Hungarian power plants using 500 km radius deposition fields and HySPLIT model.
CHAPTER 4: DISCUSSION

Using this approach in LCA would be viable if much more country would have their CTPs. This will require more work. However, the proposed approach is applied using one country. It is expected that my conclusions hold true for many other countries.

In Hungary the wind directions are moderately distributed, however there are places in Earth where the wind blows mainly from one direction all the year around. In this case the De Caritat type model would loose from its relevance, while Gaussian-type, which results a seemingly distorted deposition area shape along main wind directions, would be more applicable.

According to Heijungs (2013), regionalised LCIA can loose the connection with the whole LCA framework, but from an other viewpoint this phenomenon can be seen as a process of approaching different environmental assessment methods. Hence, it is important to emphasize that the method can be used in other applications such as chemical risk assessment or environmental impact assessment or any other method where conversion of an array of emissions of different substances to aggregated indicators is necessary.

4.1 LIMITATIONS

A very important limitation of the method is that does not take a high fraction of the emitted mass into account. Uncertainties associated with predictions of the models are increasing with the distance from the source. To solve this problem, an European-specific averaged CTP could be introduced for the remaining mass, which is likely to deposit in a larger scale.

Little is known how the CaCO$_3$ content affects CTPs of Cu in soil. This limitation does not change the conclusions about the effect of weighting, because the number of the soils for CaCO$_3$ above 5% is below 10% in the deposition areas.
• It is clearly showed that averaging across deposition area reduces geographic variability in the CTP of Cu in case of airborne emissions. Something similar has been already observed for acidifying emissions (Posch & Seppälä 2008), but was not quantified for metals.

• However, for countries where soils are relatively heterogeneous (in terms of pH and organic carbon content), such as in Hungary and neighboring regions, averaged CTP will be more or less equal irrespectively of plant location. This may not be a case in countries where there are large gradients in pH (for example, east Spain with alkaline soils against west Spain with acidic soils).

• In this specific case often there are larger differences in the weighted CTP between different models than between different locations. If the variability of the pH and organic carbon content is heterogenous, such as in the case of Hungary, a simple exponential decline function like the one developed by De Caritat is sufficient, however it needs more research to adjust the model and experimental data to validate it. Hysplit gives more realistic results however it needs more experience and it is much harder to investigate the sensitivities towards input parameters, because it has many built-in algorithms hidden away from its users. The Gaussian seems to be a promising optimum solution however it should be revised in order to reduce sensitivity towards wet deposition.

• Gaussian type model systematically estimates lower CTPs compared to the other two models. In case of Hungary it the reason probably comes from the geographic position of Hungary in the Carpathian basin, namely a relatively low plain surrounded by mountains. In this case the precipitation can be a relevant factor in soil pH, as one can see can see in Map 2, so the middle of Hungary has pH around 7-8, while the outer areas around 5-6. Thus when a model gives higher weight to further areas (e.g. HYPLIT), in this specific case, the area average pH can be lower, which would influence the area weighted CTP as well.
Citations


Hauschild, MZ & Goedkoop, M., 2013. Identifying best existing practice for characterization modeling in life cycle impact assessment. *International Journal of...


How big is small enough? Methodology and case study of electricity generation.  


APPENDIX

1 TECHNICAL DETAILS OF CALCULATION AND MODELING (REFERS TO SECTION 2.2 - 2.4).

1.1 Introduction

In the following section, a step-by-step description is provided about the modeling, from obtaining and preparing different data sources to the implementation of equations and calculations involved in the thesis.

In the next pages - for easier reading - the MATLAB code has been highlighted in 'code snippet' boxes with the default colors and font type as in MATLAB 2013a:

MATLAB code snippet example

1.2 Export data from Harmonized World Soil Database (HWSD) and Load it to MATLAB (refers to Section 2.2.2)

For calculation reasons, the extent of the original (16000x32000) global raster has been reduced to cover more or less the area of Europe. It could be done in ArcCatalog (right click/export/different data format, then in the 'Environments' subpage set the 'Processing extent' as Display). Then it has been exported to an ASCII file via 'Raster to ASCII' tool (ArcToolbox/Conversion Tools/From Raster/Raster to ASCII). This text file can be directly loaded into Matlab by its arcgridread function.

In parallel, using the HWSD's MS Access database, the necessary fields (topsoil pH, clay content, organic carbon content) can be extracted in Access with a crosstab query wizard (settings: Table: HWSD data; row heading: MU_GLOBAL; column heading: SEQ, intersection: Share, T-OC, T-Clay, T-CaCO3, T-PH). We exported the resulting table to excel, and calculated the weighted average from from the sequences. Then it could also be loaded to MATLAB as variable hwsdattr.

An 'arcgridload' script has been written to ease the import of HWSD grid and its attribute table to the MATLAB environment. It specifies the filename from which the export
starts, then checks if hwsdattr has been already imported:

```matlab
hwsdfile = 'hwsd_euro.txt';
if exist('hwsdattr.mat','file')==2
    load('hwsdattr.mat')
else
    error('HWSD attribute table needs to be imported from spreadsheet first (right click on .xlsx file, then Import data)')
end
if exist(hwsdfile,'file')==2
    [hwd, hwsdref] = arcgridread(hwsdfile);
else
    error('The specified HWSD data file does not exist.')
end
save ('hwsd.mat', 'hwsd', 'hwsdref', 'hwsdattr')
```

The 'hwsdref' variable is a referencing matrix, containing all the important georeferencing information (namely the raster cell size, and the geographical coordinates of the northwestern corner; more information can be found in its online help).

### 1.3 The 'model.mat' script - input parameter setup

Clear workspace, then set up linear regression coefficients for calculation of CTP's in soils (Owsianiak et al. 2013):

```matlab
clear all
a=5.653;
b=-0.408;
c=1.150;
```

First the most important data needs to be given: the area of the calculation (set by the 'rad' variable radius of circle around source in [km], deposition model type ('decaritat', 'hysplit',or 'gaussian') and the emission location (latitude, longitude) in [degrees]

```matlab
rad = 200;
type = 'gaussian';
location = [46.08333,18.2333];
```

Each deposition model needs different input parameters, so each case has been handled separately. Parameters for Gaussian type model (see Section 2.4.4) begins with important stack and emission source parameters and meteorological settings. Note that the windrose (wind direction distribution) data is in percent and starts from North going clockwise.

---

3 [http://www.mathworks.se/help/map/ref/makerefmat.html](http://www.mathworks.se/help/map/ref/makerefmat.html), access date: 09.04.2013
Contaminant parameters:

\[
\begin{align*}
\rho &= 1000; \quad \text{% density of particles (kg/m}^3) \\
R &= \{0.2e-6, 1.5e-6, 6e-6, 14e-6, 40e-6\}; \quad \text{% diameter of PM particles (m).} \\
V &= \{0.00065, 0.0025, 0.0071, 0.0132, 0.067\}; \quad \text{% Dry deposition} \\
\text{velocity} \\
Wr &= \{5.05e4, 3.78e5, 3.78e5, 3.78e5, 3.78e5\}; \quad \text{% scavenging ratios}
\end{align*}
\]

The 'cell' variable sets the size of one side of one raster point:

\[
cell = 10000; \quad \text{% raster point size (m)}
\]

Less input needs the model proposed by (de Caritat 1997) (refers to Section 2.4.2):

\[
\begin{align*}
case \ 'decaritat' \\
k &= -2.0031; \\
d1 &= 14220.67; \\
d0 &= 0; \\
x_c &= 0.2;
\end{align*}
\]

For HYSPLIT model, only the file name of exported result has been set (refers to Section 2.4.3):

\[
\begin{align*}
\text{case } \ 'hysplit' \\
filename='cdump.txt';
\end{align*}
\]

1.4 Comparative toxicity potentials of soils based on HWSD grid (refers to Section 2.2)

The next step is the loading or creating CaCO\textsubscript{3}, clay, organic carbon content, and pH rasters by joining the exported Harmonized Word Soil Database (HWSD) and its appropriate attribute table.

If the location of emission is the same as in the previous run we can spare this step, else
load the exported hwsd raster with its attributes:

```matlab
if exist('prevloc.mat','file')==2
    load('prevloc.mat')
else
    prevloc = [];
end
if isequal([location,rad],prevloc)==0 || exist('ctpgrid.mat','file')~=2
    if exist('hwsd.mat','file')==2 % Check if hwsd exists
        load('hwsd.mat') % Loading HWSD grid variables
    else
        error('HWSD raster needs to be imported [...]')
    end
end
```

Check if 'layers.mat' file exists then load, else create the layers:

```matlab
if exist('layers.mat','file')==2
    load('layers.mat') %loading layers
else
    clay=zeros(size(hwsd))/0; %create NaN matrices
    ph=clay;
    orgc=clay;
    caco3=clay;
end
```

Join the attribute table with HWSD grid, than saving the layers as thematic map grids (see Fig. 1):

```matlab
for i=1:length(hwsdata(:,1))
    [row, col] = find (hws == hwsdata(i,1));
    clay(sub2ind(size(hwsd),row, col)) = hwsdata(i,2);
    ph(sub2ind(size(hwsd),row, col)) = hwsdata(i,3);
    orgc(sub2ind(size(hwsd),row, col)) = hwsdata(i,4);
    caco3(sub2ind(size(hwsd),row, col)) = hwsdata(i,5);
end
save ('layers.mat','orgc','clay','caco3','ph')
clearvars row col i;
end
```
The layers covering Europe are ready at this point, but its extent is still much larger than
the necessary so the next thing to do is to cut out the necessary area defined by the
radius around emission source. Using MATLAB Mapping Toolbox function `scircle1` the
coordinates of the points of a circle around emission location can be calculated. The
inputs are the coordinates of the single point source (variable `location`) and radius of the
circle (rad).

\[
\text{[latc, longc]} = \text{scircle1(location(1), location(2), km2deg(rad))};
\]

To set up the new raster size (variable `gridsize`) and to define the new georeferencing
matrix the northeastern corner can be obtained by finding the maximum latitude and
minimum longitude coordinates in the circle previously created.

\[
\begin{align*}
\text{px} &= \text{hwsdref}(2,1); \quad \% \text{ read cell size (in degrees)} \\
\text{gridsize} &= \text{round}([(\text{max(latc)}-\text{min(latc)})/\text{px}, (\text{max(longc)} \ldots \\
&\quad -\text{min(longc)})/\text{px}]); \\
\text{corner} &= \text{round}([(\text{hwsdref}(3,2)-\text{max(latc)})/\text{px}, (\text{min(longc)} \ldots \\
&\quad -\text{hwsdref}(3,1))/\text{px}]); \\
\text{phgrid} &= \text{ph(corner(1):corner(1)+gridsize(1),corner(2):corner(2) \ldots} \\
&\quad +\text{gridsize(2))}; \\
\text{orgcgrid} &= \text{orgc(corner(1):corner(1)+gridsize(1),corner(2): \ldots} \\
&\quad \text{corner(2) +gridsize(2))}; \\
\text{save ('grids.mat','orgcgrid','phgrid')} \\
\text{ctpref} &= \text{[hwsdref(1:2,1:2); min(longc), max(latc)]};
\end{align*}
\]

Finally the calculation of CTP’s per soil unit follows using the formula by (Owsianiak et al.
2013), then clear variables:

\[
\begin{align*}
\text{ctpgrid} &= a + b.*\text{phgrid} + c.*\text{orgcgrid}; \\
\text{save ('ctpgrid.mat','ctpgrid','ctpref')}
\end{align*}
\]
1.5 Calculate distance and area matrix

In the so-called 'distance' matrix each cell has the value of geographic distance from the emission source in [km], and 'area' matrix, where the raster points have and later we make use of this two type of raster.

```matlab
if isequal(prevloc, location) == 1 &&
    exist('dist.mat','file')==2
    load('dist.mat')
else
    dist= ctpgrid;
    for i=1:gridsize(1);
        for j=1:gridsize(2);
            r = [ctpref(3,2)+((-i+0.5)*px), ctpref(3,1)+((j-0.5)*px)];
            dist(i,j) = distance(r(1),r(2), location(1), location(2));
        end
    end
    dist = deg2km(dist); % set unit to km
    dist(dist>200) = NaN; % set distance to NaN out of the circle
    save ('dist.mat','dist')
end
```

1.6 Deposition (HYSPLIT)

Since the HYSPLIT model has its own application, the MATLAB script is limited to the import of the HYSPLIT results and convert them to an appropriate grid for later calculations and display. It checks if the file exists then open it read its columns according to format string:

```matlab
switch(type)
    case 'hysplit'
        if exist('hysplit.mat','file')~=2
            if exist(filename,'file')~=2
                error('No Hysplit file has been found...[']
            else
                fileID = fopen('cdump.txt','r');
                dataArray = textscan(fileID, '%s%s%s%f%f%f%f%f%f
%...[^\n\r]', 'Delimiter', ',', 'HeaderLines', 1, ... 'ReturnOnError', false);
            fclose(fileID);
```
Cut coordinates off of table and sum of deposited particle groups

```matlab
hysplitraw = [dataArray{1:end-1}];
hysplitline = sum(hysplitraw(:,3:end),2);
```

Create grid from vectorized data (if the HYSPLIT run set correctly there are the same, 'N' number of rows and columns):

```matlab
N = sqrt(numel(hysplitline));
hysplit = flipud(reshape(hysplitline,N,N));
ref = [max(hysplitraw(:,1)) min(hysplitraw(:,2))];
clearvars hysplitline hysplitraw fileID dataArray ans;
end
else
 load('hysplit.mat')
end
```

### HYSPLIT result table

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Longitude</th>
<th>Particle group 1</th>
<th>Particle group 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>43.5</td>
<td>5.4</td>
<td>3.00E-010</td>
<td>7.70E-009</td>
</tr>
<tr>
<td>43.5</td>
<td>5.5</td>
<td>2.80E-008</td>
<td>1.11E-008</td>
</tr>
<tr>
<td>43.6</td>
<td>5.4</td>
<td>2.80E-008</td>
<td>3.20E-009</td>
</tr>
<tr>
<td>43.6</td>
<td>5.5</td>
<td>7.40E-009</td>
<td>2.30E-007</td>
</tr>
<tr>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

### Deposition raster

<table>
<thead>
<tr>
<th>X</th>
<th>Y</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.00E-090</td>
<td>3.91E-008</td>
</tr>
<tr>
<td>3.12E-008</td>
<td>2.37E-007</td>
</tr>
<tr>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

Cut out the necessary area, using the northwestern corner cell of the new grid:

```matlab
corner = round(([ref(1)-ctpref(3,2))/px, (ctpref(3,1) ... -ref(2))/px));
dep = hysplit(corner(1):corner(1)+gridsize(1)-1,corner(2) ... :corner(2)+gridsize(2)-1);
```

1.7 Deposition (Gaussian)

Set up various parameters for the atmospheric dispersion:

```matlab
    case 'gaussian'
        size.n = numel(R);  % number of particle size group
        dt = 1 /numel(U);   % one season or month (in seconds)
        dtwet = P/ J/365*24 / numel(U);  % wet deposition time (s)
        dtdry = dt-dtwet;  % dry deposition time (s)
        rad = rad*1000;   % change unit from km to m
```
Interpolating windrose to higher resolution, where ‘\texttt{winddir}’ is the number of windrose directions in the original data, and N is interpolation factor (to how much parts each sector is further divided):

\begin{Verbatim}
winddir= numel(windrose);
N=10;
hrwindrose = zeros(1,(N+1)*winddir);
for k = 1:(winddir-1)
    if windrose(k) == windrose(k+1)
        hrwindrose(k*(N+1)-N:k*(N+1)) = ...
        windrose(k)*ones(1,N+1);
    else
        hrwindrose(k*(N+1)-N:k*(N+1)) = windrose(k):...
        (windrose(k+1)-windrose(k))/(N+1):
        windrose(k+1)-
        windrose(k))/(N+1));
    end
end
\end{Verbatim}

If two neighbouring wind direction are the same then insert the same data N times between them, else put data points equally distributed between two neighbouring wind direction:

\begin{Verbatim}
if windrose(end)==windrose(1)
    hrwindrose(winddir*(N+1):N:winddir*) = ...
    windrose(1)*ones(1,N+1);
else
    hrwindrose(winddir*(N+1)-N:winddir*(N+1)) = windrose(1):...
    (windrose(1)-windrose(winddir))/(N+1):
    windrose(1)-
    windrose(winddir)/(N+1));
end
\end{Verbatim}

Interpolating between last and first angle and rescale to \([0,1]\) interval:

\begin{Verbatim}
if windrose(end)==windrose(1)
    hrwindrose(winddir*(N+1)-
    N:end)=windrose(1)*ones(1,N+1);
else hrwindrose(winddir*(N+1)-N:end) =
    windrose(winddir):...
    (windrose(1)-
    windrose(winddir))/(N+1):windrose(1)... 
    -(windrose(1)-windrose(winddir))/(N+1));
    hrwindrose = hrwindrose/N/100;
end
\end{Verbatim}

Set up parameters for calculating settling velocity:

\begin{Verbatim}
g  = 9.81;  % \texttt{gravitational acceleration (m/s}^2)\texttt{)}
mu    = 1.8e-5;  % \texttt{dynamic viscosity of air (kg/m.s)}
shape = 1.0;  % \texttt{shape factor}
freep = 6.53e-8;  % \texttt{free main path in air}
rhoair = 1.2754;  % \texttt{air density (kg/m}^3\texttt{)}
W  = 2.*rho.*g.*R.^2 ./ (9.*mu);  % \texttt{settling velocity (m/s)}
frea = freep* (rho/rhoair);  % \texttt{slip correction factor}
\end{Verbatim}

Applying Cunningham slip factor for particles smaller than 15 um:
for k = 1:numel(R)
    if R(k) < 15e-6
        sc = 1+(2*frea/2*R(k))*(1.26+0.4*exp(-0.55*2*R(k)/frea));
        W(k)=W(k)*sc/shape;
    end
end

Set receptor grid parameters:

A = cell^2; % receptor area (one raster point) (m^2)
xlim = [-rad, rad];
ylim = [-rad, rad];
[x, y] = meshgrid(-rad:cell:rad,-rad:cell:rad); % mesh points

Calculate ground-level PM deposition flux from the source at all grid points using Ermak's solution, shifting the (x,y) coordinates so the source location is at the origin, then scale the result by (A*dt) to obtain a total deposition (in MU) over the time interval 'dtdry' and 'dtwet' for dry and wet deposition, respectively:

dep = 0;
depdir = 0;
depsize = 0;
J= J/(1000*3600);
for j = 1 : size.n, %run for all particle size group
    depsize = (dtdry *V(j)*ermak(x,y,0,h,U,V(j),W(j),class,...
        'dry', lambda ))+
        (dtwet*J*Wr(j)*ermak(x,y,0,h,U,V(j),W(j),... class, 'wet', lambda ));
end

Rotate plume for each direction, and weight deposition by wind direction distribution:

for k = 1 :(winddir*(N+1)),
    depdir = depdir + hrwindrose(k)*imrotate(depsize,...
        90-k*(360/(winddir*(N+1))),'nearest','crop');
end
dep = dep + depdir;

Transform deposition field to geographic extent, and change unit from [MU m^{-2}] to [MU km^{-2}]

dep = imresize(dep,gridsize);
dep = 1e6*dep;
rad = rad/1000;
1.8 Deposition by de Caritat

```matlab
case 'decaritat'
    dep = d1.*(dist.^k);  % "outside" function
    dep (dep>d1) = NaN;
end
```

Save location for the next runs, and clear workspace

```matlab
prevloc = [location, rad];
save ('prevloc.mat', 'prevloc')
clearvars -EXCEPT dep ctpref ctpgrid cu type dist;
```

1.9 Weighting CTPs

```matlab
dep(isnan(dep))=0;
while sum(sum(area.*dep*1e6))>1 %reduce deposition area to include
    only emitted mass
    dist(dist>(max(max(dist))-10))= NaN; %reduce the circle by 10 kms
    dep = -(dep.*(isnan(dist)-1)); %set deposition to zero outside
    circle
%end
weighted = ctpgrid.*landuse;
```

1.10 Code of Ermak’s function

The deposition script has been implemented as a function script to enable sequential
running of it (I used the script provided in [http://people.math.sfu.ca/~stockie/atmos/](http://people.math.sfu.ca/~stockie/atmos/)):

```matlab
function F = ermak( x, y, z, H, U, W, V, stability)
```

Input parameters:
- X - receptor locations: distance along the wind direction, with the source at
  x=0 (m)
- Y - receptor locations: cross-wind direction (m)
- Z - receptor locations: vertical height (m)
- H - source height (m)
- U - wind velocity (m/s)
- W - gravitational settling velocity (m/s)
- V - deposition velocity (m/s)
- stability - Pasquill-Guifford stability class ('A'-'F')
F - contaminant deposition (MU/m^2/s)

Set dispersion coefficients, and wind speed power law parameter 'p' using Pasquill-Gifford curves:

```matlab
switch(stability)
    case 'A'
        p = 0.7;
        coeffs_y=[24.1670, 2.5334];
        coeffs_z= [0.10 122.800 0.94470; 0.16 158.080 1.05420; ...
                   3.11 453.850 2.11660; inf nan nan];
        case 'B'
        ...
        case 'C'
        [... and D, E, F classes...]
    otherwise
        error('gaussianPlume:stability', [Unknown ...
                                        stability class ', stability]);
end
```

Construct sigma_y vector along the x-axis:

```matlab
n=1;
while x/1000 > coeffs_z(n,1)
    n=n+1;
end
```

Determine the sigma coefficients based on stability class:

```matlab
bz = coeffs_z(n,3);
az = coeffs_z(n,2);
sigmay = 465.11628.*(abs(x./1000)).*tan(0.017453293.*(coeffs_y(1)... -coeffs_y(2) .*log(abs(x./1000)))).* (x>0);
sigmaz = az*abs(x./1000).*bz .* (x > 0);
```

Clip all values greater than 5e3 for stability classes A-C:

```matlab
switch(stability)
    case {'A', 'B', 'C'}
        if sigmay>5e3
            sigmay=5e3;
        end
end
```
Eddy diffusivity $[m^2 s^{-1}]$:

$$K_z = 0.5 \cdot (\text{sigmaz}.^2) \cdot U./\text{abs}(x).\cdot (x > 0);$$

Calculate the contaminant concentration (kg m$^{-3}$) using Ermak's formula:

$$Wo = V - 0.5*W;$$

$$F = \frac{1}{(2\cdot\pi\cdot U_{eff}\cdot \text{sigmay}\cdot \text{sigmaz})}\cdot \exp\left(-0.5*W^2/\text{sigmay}.^2\right)\cdot \exp\left(-0.5*W^2/\text{Kz}^2\right)\cdot \exp\left(-0.5*(z-H)/Kz\cdot 8/\text{Kz}^2\right)\cdot \exp\left(-0.5*W^2/\text{sigmaz}.^2\right)\cdot \exp\left(-0.5*W^2/\text{Kz}\cdot \text{W}^2/\text{sigmaz}.^2\right)\cdot \text{erfc}\left(\text{Wo}\cdot \text{sigmaz}/\sqrt{2}/\text{Kz} + (z+H)/\sqrt{2}/\text{sigmaz}\right);$$

2 DATA TABLES

Table 1. Estimates of parameters in eq. (2,3) for six stability class and urban terrain

<table>
<thead>
<tr>
<th>Stability class</th>
<th>x (km)</th>
<th>a$_z$</th>
<th>b$_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.1</td>
<td>122.8</td>
<td>0.94</td>
</tr>
<tr>
<td></td>
<td>0.16</td>
<td>158.08</td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>0.21</td>
<td>170.22</td>
<td>1.09</td>
</tr>
<tr>
<td></td>
<td>0.26</td>
<td>179.52</td>
<td>1.13</td>
</tr>
<tr>
<td></td>
<td>0.31</td>
<td>217.41</td>
<td>1.26</td>
</tr>
<tr>
<td></td>
<td>0.41</td>
<td>258.89</td>
<td>1.41</td>
</tr>
<tr>
<td></td>
<td>0.51</td>
<td>346.75</td>
<td>1.73</td>
</tr>
<tr>
<td></td>
<td>3.11</td>
<td>453.85</td>
<td>2.12</td>
</tr>
<tr>
<td></td>
<td>&gt;3.11</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>B</td>
<td>0.2</td>
<td>90.67</td>
<td>0.93</td>
</tr>
<tr>
<td></td>
<td>0.4</td>
<td>98.48</td>
<td>0.98</td>
</tr>
<tr>
<td></td>
<td>&gt;0.4</td>
<td>109.3</td>
<td>1.1</td>
</tr>
<tr>
<td>C</td>
<td>61.14</td>
<td>0.91</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>0.31</td>
<td>34.46</td>
<td>0.87</td>
</tr>
<tr>
<td></td>
<td>1.01</td>
<td>32.09</td>
<td>0.81</td>
</tr>
<tr>
<td></td>
<td>3.01</td>
<td>32.09</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>10.01</td>
<td>33.5</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>36.65</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td>&gt;30</td>
<td>44.05</td>
<td>0.51</td>
</tr>
<tr>
<td>E</td>
<td>0.1</td>
<td>24.26</td>
<td>0.84</td>
</tr>
<tr>
<td></td>
<td>0.31</td>
<td>23.33</td>
<td>0.82</td>
</tr>
<tr>
<td></td>
<td>1.01</td>
<td>21.63</td>
<td>0.76</td>
</tr>
<tr>
<td></td>
<td>2.01</td>
<td>21.63</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>4.01</td>
<td>22.53</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td>10.01</td>
<td>24.7</td>
<td>0.51</td>
</tr>
<tr>
<td></td>
<td>20.01</td>
<td>26.97</td>
<td>0.47</td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>35.42</td>
<td>0.38</td>
</tr>
<tr>
<td></td>
<td>&gt;40</td>
<td>47.62</td>
<td>0.3</td>
</tr>
<tr>
<td>F</td>
<td>0.21</td>
<td>15.21</td>
<td>0.82</td>
</tr>
<tr>
<td></td>
<td>0.71</td>
<td>14.46</td>
<td>0.78</td>
</tr>
<tr>
<td></td>
<td>1.01</td>
<td>13.95</td>
<td>0.68</td>
</tr>
<tr>
<td></td>
<td>2.01</td>
<td>13.95</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>3.01</td>
<td>15.01</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>4.01</td>
<td>15.01</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>10.01</td>
<td>30.01</td>
<td>0.42</td>
</tr>
<tr>
<td></td>
<td>20.01</td>
<td>30.01</td>
<td>0.33</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>36.65</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td>&gt;30</td>
<td>44.05</td>
<td>0.51</td>
</tr>
<tr>
<td></td>
<td>&gt;60</td>
<td>34.22</td>
<td>0.22</td>
</tr>
</tbody>
</table>
Table 2. Emission location specific annual average meteorological data used in Gaussian type atmospheric model

<table>
<thead>
<tr>
<th>Power plant</th>
<th>Average wind speed</th>
<th>wind direction</th>
<th>Annual precipitation (P) [mm]</th>
<th>Observation point</th>
</tr>
</thead>
<tbody>
<tr>
<td>Borsod</td>
<td>3</td>
<td>[16,8,6,10,17,5,17,20]</td>
<td>1139</td>
<td>Miskolc</td>
</tr>
<tr>
<td>Vértesi</td>
<td>3</td>
<td>[8,4,11,14,11,10,14,29]</td>
<td>912</td>
<td>Győr</td>
</tr>
<tr>
<td>Ajka</td>
<td>3</td>
<td>[22,4,4,8,29,10,6,18]</td>
<td>935</td>
<td>Pápa</td>
</tr>
<tr>
<td>Tiszai</td>
<td>3</td>
<td>[16,8,6,10,17,5,17,20]</td>
<td>1030</td>
<td>Miskolc</td>
</tr>
<tr>
<td>Mátrai</td>
<td>3.3</td>
<td>[12,15,9,7,14,11,18,12]</td>
<td>997</td>
<td>Szolnok</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Power plant</th>
<th>Latitude (degree)</th>
<th>Longitude (degree)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mátrai</td>
<td>47.789759</td>
<td>20.066722</td>
</tr>
<tr>
<td>Vértesi (Oroszlány)</td>
<td>47.486706</td>
<td>18.31225</td>
</tr>
<tr>
<td>Tiszapalkonya</td>
<td>47.92291</td>
<td>21.052195</td>
</tr>
<tr>
<td>Borsod</td>
<td>48.24488</td>
<td>20.669235</td>
</tr>
<tr>
<td>Ajka</td>
<td>47.1068802</td>
<td>17.563957</td>
</tr>
</tbody>
</table>