Preface

After the first year of lectures and a first research project, master-students at the center for energy and environmental studies, IVEM, perform their final research project. This report presents the outcome of such a project. The project represents 32 ECTS-credits (European Community Course Credit Transfer System), which corresponds to 896 hours.

I performed this research at the Coordination Center for Effects (CCE) at the Netherlands Environmental Assessment Agency (Dutch: NMP, Natuur- en MilieuPlanburau). NMP is part of National Institute for Public Health and the Environment (Dutch: RIVM, Rijksdienst voor Volksgezondheid en Milieu) situated in Bilthoven. Jean-Paul Hettelingh (CCE) coached me during the project. His long experience with integrated assessment and air pollution policy was very helpful. I want to thank him for his professional and inspiring support. Also Jan Aben (NMP) was involved in the project. His major contributions related to the application of Eutrend, but he gave many other useful comments as well. For example, Figure 7.1 is based on his suggestions. Thank you, Jan for your skilful support and kind cooperation. From the IVEM José Potting supervised and initiated the project. Thank you José for introducing me into the world of environmental modelling. René Benders served as second supervisor. He had to judge the project since José was unavailable at that time. Thanks René for your flexibility and willingness to take over the supervision at such short notice.

At CCE I enjoyed the company of Max Posch, Jaap Slootweg, and Maarten van 't Zelfde. They are great people. Max helped me with all possible computer programs. He didn’t get irritated after I exchanged the horizontal and vertical coordinates of the EMEP grid, for the second time. Together with Jean-Paul and Jan these people formed the supportive environment in which I was allowed to do my project. Thank you all.

Sandra Bellekom, July 2005.
# Table of contents

Summary ........................................................................................................................................... 7

Samenvatting ..................................................................................................................................... 9

1 Introduction .......................................................................................................................................... 11
1.1 Acidification ................................................................................................................................. 11
1.2 The Convention ............................................................................................................................ 11
1.3 Use of acidification factors .......................................................................................................... 16
1.4 Problem setting ............................................................................................................................ 18
1.5 Research objective ....................................................................................................................... 19
1.6 Research questions ....................................................................................................................... 19
1.7 The aggregation problem ............................................................................................................ 19
   1.7.1 Nearly decomposable systems, hierarchy, and the aggregation problem ............................... 19
   1.7.2 Models and aggregation ........................................................................................................ 20
   1.7.3 Aggregation error ................................................................................................................ 22
   1.7.4 Spatial resolution ................................................................................................................ 23
1.8 Structure of the report ................................................................................................................... 24

2 Approach ............................................................................................................................................ 25
2.1 Boundary settings ......................................................................................................................... 25
2.2 Creation of $\Delta A(\Delta E)$ graphs ............................................................................................... 25
   2.2.1 Emission values ..................................................................................................................... 26
   2.2.2 Emission sources ................................................................................................................ 26
   2.2.3 Dispersion and deposition .................................................................................................... 26
   2.2.4 Access critical load database .............................................................................................. 28
   2.2.5 $\Delta A(\Delta E)$ calculations .................................................................................................. 29
2.3 Analysis of $\Delta A(\Delta E)$ .............................................................................................................. 29
   2.3.1 Variability of AF .................................................................................................................. 30
   2.3.2 Value of the acidification factor .......................................................................................... 31
   2.3.3 Linearization error .............................................................................................................. 34

3 Results from spatial emission/deposition resolution analysis ......................................................... 39
3.1 $\Delta A(\Delta E)$ graphs .................................................................................................................... 39
   3.1.1 Location of ecosystems changing their state .......................................................................... 40
3.2 Variability of AF .......................................................................................................................... 45
3.3 Value of AFs ................................................................................................................................. 47
3.4 Error ............................................................................................................................................ 48
3.5 Aggregated deposition method ................................................................................................... 50
   3.5.1 $\Delta A(\Delta E)$ graphs ........................................................................................................... 51
   3.5.2 Variability of AF .................................................................................................................. 52
   3.5.3 Values of AFs ..................................................................................................................... 52
   3.5.4 Error ................................................................................................................................... 53
3.6 Summary and conclusions ............................................................................................................ 53

4 Results from sector specific emission changes ............................................................................. 55
4.1 $\Delta A(\Delta E)$ graphs .................................................................................................................... 55
4.2 Value of AFs ................................................................................................................................. 57
4.3 Error ............................................................................................................................................ 59
4.4 Summary and conclusions ............................................................................................................ 61
Summary

Acidification is still an important environmental burden that needs attention. Even nowadays large parts of central Europe (e.g. the Netherlands, Germany, and Poland) are heavily acidified.

In this study European acidification, $A$, is expressed as “area of unprotected ecosystem in Europe”. An ecosystem is an area with uniform soil, vegetation, and organisms, like a forest or grassland. “Protected” and “unprotected” are terms used to indicate whether or not an ecosystem is harmed by the acidifying deposition it receives. When the $SO_2$, $NO_x$, and $NH_3$ deposition on an ecosystem exceed the so called critical load of that ecosystem, the ecosystem is “unprotected”. Otherwise it is “protected” against acidification. This approach enables a quantification of the environmental effect acidification.

Extended (integrated assessment) models determine acidification, $A$, resulting from emissions originating in European countries. However, these models require much computing time. Traditionally, acidification factors (AFs) linearly relate national emission changes ($\Delta E$) to changes in European acidification ($\Delta A$): $\Delta A = AF \cdot \Delta E$. For each country and each substance a separate AF exists. AFs can fasten acidification calculations, enabling for example real time computations during negotiations on emission reductions.

Life cycle assessment (LCA) uses AFs to determine the acidifying impact from a product or service. AFs should approximate acidification with a reasonable accuracy and should be generally applicable. They should not depend heavily on model parameters like resolution.

This report deals with the research question: how do AFs depend on spatial resolutions of emission and deposition, sector specific emissions, and grid cell specific emissions. We also try to indicate which kind of AFs, considering calculation method and $\Delta E$ range, are to be preferred (considering the previously listed model parameters).

Emissions and deposition resolution specific AFs relate $\Delta E$ from one country to $\Delta A$ in Europe. These national AFs are calculated with different emission and/or deposition resolutions of the extended model. Sector specific AFs relate $\Delta E$ from one economic sector within a country to $\Delta A$ in Europe. Using the national AFs instead of sector specific AFs will probably produce less accurate results when studying the influence of emission reduction within just one economic sector. Grid cell specific AFs relate $\Delta E$ from a certain grid cell of $50x50km^2$ to $\Delta A$ in Europe. Such AFs can be used to accurately determine the acidifying impact of emissions from a specific location (or small area).

The current study considers AFs resulting from two different calculation methods and three $\Delta E$ ranges. Ideally AFs do not depend on model parameters. A low sensitivity of AFs to emission/deposition resolution, sector specific emission, and grid cell specific emissions is preferable. The linearization error related to applying AFs instead of the extended model is important as well. Not only is a low value of the error wanted for optimal AFs but also a low sensitivity of the error to the model parameters.

To answer the research questions we calculate $\Delta A$ for given $\Delta E$, using different emission and deposition resolutions in the extended model. $\Delta E$ ranges from -50% to +20% with steps of 1%. AF equals the slope of a straight line through these points.

In general it turned out that AFs depend in a non-systematic way on emission and deposition resolutions. Variations in AFs and linearization errors strongly correlate to the occurrence and size of so called “jumps”. Each $\Delta E$ step of 1% causes a step in $\Delta A$. Some $\Delta A$-steps are considerably larger than average; these are called “jumps”. The size of $\Delta A$-
steps is determined by the number of ecosystems changing their state from “protected” to “unprotected” (or the other way around) and the area of these ecosystems. A step size of zero occurs when the change in deposition on the European ecosystems does not cause a change of the state of any of these ecosystems. This happens when deposition reductions (resulting from emission reductions) are too small to flip an ecosystem from “unprotected” to “protected”, when ecosystems are highly exceeding their critical load and requiring a very large deposition reduction to change their state, or when the ecosystems were already “protected”. When the $\Delta A$-step is not zero, the minimum situation is that one ecosystem changes its state. In that situation the $\Delta A$-step equals the area of this single ecosystem. The current research showed that often “jumps” are mainly caused by just one large ecosystem changing its state. Therefore, a smaller maximum size of ecosystems would result in smaller variations in AFs and smaller linearization errors.

Both, sector and grid cell specific AFs deviate from the national AFs. This deviation is caused by a number of aspects including different absolute emission amounts. Grid cell specific AFs could not be calculated in a satisfying way applying the used model, because of the very small emission amounts involved. Large ranges of grid cell specific $\Delta E$ (expressed as percentage of the grid cell specific emission) caused zero sized $\Delta A$-steps.

For all experiments two methods to compute AFs were applied. AFs calculated using regression and using the Single Value (SV) method were compared. The SV method defines AF as $\Delta A/\Delta E$ at a certain point, we use $\Delta E = -50\%$. Regression techniques calculate a straight line through the points while minimizing the error. Theoretically regression AFs should be less sensitive to small variations in the model results. Most experiments in this report support this. Based on theoretical and experimental results we recommend regression AFs because they depend less on the studied model parameters.

Regression AFs are computed over a certain range of emission changes. The report distinguishes regression AFs related to the whole emission range ($-50\% \leq \Delta E \leq +20\%$), large emission reduction mainly used in air pollution policy application ($-50\% \leq \Delta E \leq -20\%$), and small emission changes mainly appearing in LCA applications ($-20\% \leq \Delta E \leq +20\%$). We expected the emission range specific AFs to be better than the AFs for the whole emission range. This effect was only slightly observed. When considering small emission changes, the AF related to $-20\% \leq \Delta E \leq +20\%$ indeed resulted in smaller errors than the AF related to the whole emission range. However, this emission range specific AF was more sensitive to model parameters.

In LCA the emission range specific AFs could slightly improve the accuracy of the calculated acidification. More important for LCA applications is the possibility to quantify the error. Uncertainty of LCA results has received increased attention lately. However, uncertainty calculations in LCA prefer a quantification of the uncertainty in AF itself, not in the resulting acidification (as was computed in the current research). Using AFs within air pollution policy is still in its infancy. The use of AFs increases the speed of the calculation of acidification. However, the error in the calculated acidification also increases. National AFs turned out to be not generally applicable, for example for sector specific emissions, without considerably increasing the uncertainty of the calculated acidification. The report quantifies this uncertainty for some situations. Air pollution policy should weight the increased uncertainty against the gained calculation speed.
**Samenvatting**

Verzuring is een milieueffect dat nog steeds aandacht verdient. Zelfs tegenwoordig zijn grote delen van midden Europa (bijv. Nederland, Duitsland, Polen) zwaar verzuurd. In dit rapport wordt verzuring in Europa, ΔA, weergegeven als “oppervlakte onbeschermd ecosysteem in Europa”. Een ecosysteem is een gebied met gelijke grond, begroeiing en organismen, bijvoorbeeld bos of weide. “Beschermd” of “onbeschermd” wordt gebruikt om aan te geven of een ecosysteem al dan niet wordt beschadigd door de depositie die het ontvangt. Als de SO$_2$, NO$_x$, en NH$_3$ deposits de zogenaamde kritische belasting overschrijden is het ecosysteem “onbeschermd”, anders is het “beschermd” tegen verzuring. Deze aanpak maakt het mogelijk om verzuring te kwantificeren. Uitgebreide (integrated assessment) modellen berekenen de verzuring in Europa, ΔA, die ontstaat t.g.v. emissies uit Europese landen. Deze modellen vragen echter veel rekentijd. Verzuringsfactoren (engels: acidification factors, AFs) verbinden nationale emissieveranderingen (ΔE) lineair met Europese verzuring (ΔA): ΔA = AF·ΔE. Voor ieder land en voor iedere stof (SO$_2$, NO$_x$, en NH$_3$) bestaat een eigen AF. AFs versnellen de berekening van verzuring waardoor het bijvoorbeeld mogelijk wordt om tijdens onderhandelingen over emissieverlagingen direct zulke berekeningen uit te voeren. In levenscyclusanalyse (LCA) worden AFs gebruikt om de verzuring van een product of service te bepalen. AFs moeten de verzuring redelijk nauwkeurig benaderen en ze moeten liefst algemeen toepasbaar zijn. Ze moeten niet te veel afhangen van modellparameters zoals resolutie.

Dit rapport behandelt de onderzoeksvraag: hoe hangen AFs af van de ruimtelijke resolutie van emissie en depositie, van sectorspecifieke emissies en van roostercel-specifieke emissies. We proberen ook aan te geven welke soort AFs, wat betreft berekeningsmethode en ΔE-bereik, het beste is (met inachtneming van de bovenstaande model parameters). Emissie- en depositie-resolutie-specifieke AFs relateren de ΔE van één land met de ΔA in Europa. Zulke nationale AFs worden berekend met verschillende emissie en/of depositie resoluties van het uitgebreide model. Sectorspecifieke AFs relateren de ΔE van één economische sector in een land met de ΔA in Europa. Wanneer de nationale AF i.p.v. de sectorspecifieke AF wordt gebruikt voor het berekenen van het effect van emissieverlaging in één sector, is het resultaat minder nauwkeurig. Roostercel-specifieke AFs relateren de ΔE van één specifiek roostercel van 50x50km$^2$ met de ΔA in Europa. Zulke AFs kunnen worden gebruikt om nauwkeurig de verzuring te bepalen van emissies vanuit een specifieke locatie (of klein gebied).

Het onderzoek bekijkt AFs die op twee verschillende manieren berekend zijn en het maakt onderscheidt in AFs die betrekking hebben op drie verschillende ΔE-bereiken. Idealiter hangt de AF niet af van modellparameters. We prefereren een geringe afhankelijkheid van de emissie/depositieresolutie, sector specifieke emissie en roostercel specifieke emissie. De linearisatiefout, die ontstaat bij het gebruik van AFs in plaats van het uitgebreide model, is ook belangrijk. We willen niet alleen een kleine fout maar ook een geringe afhankelijkheid van de model parameters.

Om de onderzoeksvraag te beantwoorden berekenen we ΔA voor gegeven ΔE met het uitgebreide model met verschillende emissie- en depositieresoluties. ΔE loopt van -50% tot +20% met stappen van 1%. AF is de richtingscoëfficiënt van een lijn door deze punten.

In het algemeen bleek uit ons onderzoek dat de AFs op een niet-systematische manier afhangen van de emissie- en depositieresolutie. Variaties in AFs en linearisatiefouten hangen nauw samen met het optreden van zogenaamde “sprongen”. Elke ΔE-stap van 1%
veroorzaakt een stap in $\Delta A$. Sommige $\Delta A$-stappen zijn aanzienlijk groter dan gemiddeld; deze heten “sprongen”. De hoogte van de $\Delta A$-stappen wordt bepaald door het aantal ecosysteem dat hun status verandert van “beschermd” naar “onbeschermd” (of andersom) en de oppervlakte ervan. Een stapgrootte van nul treedt op als veranderingen in depositie geen enkel Europees ecosysteem van status doen veranderen. Dit is het geval wanneer depositieverlagingen (als gevolg van emissieverlagingen) te klein zijn om een ecosysteem van “onbeschermd” naar “beschermd” om te klappen, wanneer ecosystemen hun kritische belasting ver overschrijden en dus een grote depositievermindering nodig hebben om hun status te veranderen of wanneer ecosystemen al “beschermd” waren. Als de $\Delta A$-stap geen nul is dan verandert tenminste één ecosysteem zijn status. De hoogte van de $\Delta A$-stap is dan gelijk aan de oppervlakte van dat ecosysteem. Uit ons onderzoek blijkt dat “sprongen” vaak voornamelijk veroorzaakt worden door één groot ecosysteem dan zijn status verandert. Het verkleinen van de maximale oppervlakte van ecosystemen leidt dus tot kleinere “sprongen”, wat weer resulteert in kleinere variaties in AF en kleinere fouten.

Zowel de sector- als de roostercel-specifieke AFs wijken af van de nationale AFs. Dit wordt veroorzaakt door een aantal effecten waaronder verschillen in emissiewaarden. Roostercel-specifieke AFs konden niet goed berekend worden met het model omdat de emissies vanuit één enkele roostercel vaak te klein waren om enig effect te laten zien in het model (veel $\Delta A$-stappen waren nul).

Voor alle experimenten hebben we AF op twee verschillende manieren berekend, via regressie en via de “enkele waarde” (engels: single value, SV) methode. De SV methode berekent AF als $\Delta A/\Delta E$ op een bepaald punt, wij gebruiken $\Delta E = -50\%$. Regressie is een methode om de lijn door een groep punten te bepalen zodanig dat de fout zo klein mogelijk is. Theoretisch zou de regressie-AF het minst variëren (als de modeluitkomsten variëren). De meeste experimenten in dit onderzoek laten dat ook zien. We bevelen daarom aan om regressie-AFs gebruiken en niet de AFs gebaseerd op de SV-methode. Regressie-AFs worden bepaald over een bepaald bereik van $\Delta E$. We onderscheiden het hele bereik (-50% ≤ $\Delta E$ ≤ +20%), grote emissieverminderingen zoals gebruikelijk in luchtbeleid (-50% ≤ $\Delta E$ ≤ -20%) en kleine emissieveranderingen zoals die meestal optreden in LCA (-20% ≤ $\Delta E$ ≤ +20%). De verwachting was dat emissiebereik-specifieke AFs beter zouden zijn dan de AF voor het hele bereik. Dit effect was maar een klein beetje zichtbaar in ons onderzoek. De AF voor -20% ≤ $\Delta E$ ≤ +20% gaf inderdaad iets kleinere fouten dan de AF voor het hele emissiebereik, maar deze emissiebereik-specifieke AF hing wel sterker af van de modellparameters.

Binnen LCA zou een emissiebereik-specifieke AF dus iets nauwere resultaten kunnen geven van de berekende verzuring. Belangrijker voor LCA-toepassingen is echter het kwantificeren van de fout. Onzekerheid in LCA-resultaten staat erg in de belangstelling. Echter, onzekerheidsberekeningen in LCA hebben de fout in AF zelf nodig en niet in de berekende verzuring (zoals we in dit onderzoek hebben gedaan). Het gebruik van AFs in luchtbeleid staat nog in de kinderschoenen. AFs kunnen de snelheid van de verzuringsberekeningen vergroten, maar de fout in de berekende verzuring neemt toe. Nationale AFs zijn niet algemeen toepasbaar, bijvoorbeeld voor sector-specifieke emissies, zonder dat de onzekerheid in de verzuring enorm toeneemt. Dit onderzoek heeft deze onzekerheid voor een aantal gevallen berekend. Luchtbeleid moet de grotere onzekerheid afwegen tegen de snellere berekeningen.
1 Introduction

1.1 Acidification

Acidification is the detrimental effect that acidifying substances like SO$_2$, NO$_x$, and NH$_3$ have on the environment. In the late seventies and early eighties acid rain and the threat to forests attracted a lot of attention. Thanks to international agreements the emission of acidifying substances has been decreasing since then. Figure 1.1 shows examples of national emission reduction in the Netherlands, France, and Poland for the period 1980-2002. Especially for SO$_2$, these countries realized large emission reductions.

![Figure 1.1: three examples of trends in acidifying emissions. (data from webdab.emep.int)](image)

The public’s interest in acidification has decreased. However, also nowadays acidification is a problem in Europe. Figure 1.2 shows that large parts of European forests experience a too large deposition of acidifying substances. Acidification is still an important environmental burden that needs attention.

![Figure 1.2: Percentage of forest area receiving acid deposition above the critical load in year 2000 (Amann et al., 2004).](image)

1.2 The Convention

The first visible effects of acidification appeared in the late 1960s. Fish in lakes in Scandinavian countries died. Later on also forests in Germany were damaged. The acid rains that caused these effects originated from outside the country where the effect
occurred. Because of this transboundary property, reducing the acidifying effects required international negotiations. (Tuinstra et al., 1999)

Therefore in 1979 the Convention on Long-range Transboundary Air Pollution (LRTAP) was established. “The Convention includes a process for negotiating concrete measures to control pollutants through specific agreements called protocols. It also coordinates efforts on research, monitoring and the development of emission reduction strategies on regional air pollution.” (UNECE, 2004). LRTAP produced a first air pollution protocol in 1985: the 1985 Sulphur Protocol. Countries who signed the protocol promised to reduce their national SO$_2$ emissions by 30% relative to 1980. Criticism to this protocol included: (Jost, 2004)

1. No direct relation between 30% emission reduction and ecological goals
2. Economic consequences not always known
3. Poor scientific basis

The Convention formed the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutions in Europe (EMEP) in 1983 (Tuinstra et al., 1999). EMEP provides “governments with information on the deposition and concentration of air pollutants as well as on the quantity and significance of the long-range transmission of air pollutants and fluxes across boundaries.” (Schneider & Schneider, 2004)

Figure 1.3: structure of LRTAP
Figure 1.3 shows the structure of the convention. Scientific and technical discussions take place in the task forces and other support groups, while the government officials in the working groups and EMEP steering body advice the Executive body. The Convention chooses the Regional Air Pollution Information and Simulation (RAINS) model as integrated assessment model to provide a scientific basis for the emission reduction. The lack of a scientific base was one of the points of criticism to the 1985 Sulphur Protocol.

Figure 1.4 depicts the structure of RAINS. The figure distinguishes three modules:
- OPT = optimization module
- EMCO = emission-cost module
- DEP = acid deposition and ecosystems impact module

In the following we focus on acidification. OPT determines how a given target can be reached at minimal costs. The target is formulated as deposition levels. OPT indicates which measures to reduce SO\(_2\), NO\(_x\), and NH\(_3\) emissions will decrease the emissions in such a way that the set target is reached and the costs are as low as possible. EMCO calculates the amounts of SO\(_2\), NO\(_x\), and NH\(_3\) emissions. The module also estimates the costs for emission reductions. DEP computes the deposition of emitted substances and the environmental impact thereof. Deposition maps are calculated using transfer matrices for SO\(_2\), NO\(_x\), and NH\(_3\), the so called source-receptor matrices (SRM). These SRMs originate from the EMEP long-range atmospheric transport model. They linearly relate national emissions to depositions on the EMEP-grid. Figure 1.5 depicts the considered area and the 150x150km\(^2\) EMEP grid. From the deposition maps DEP derives the acidifying impact on the environment.
As said before, one of the drawbacks of the 1985 Sulphur Protocol was the lack of a relation between emission reduction and the ecological goal. The ecological goal is to protect ecosystems against acidification. An ecosystem is an area with uniform soil, vegetation and organisms. For example forest, lakes, wetland, grassland, and shrubs are distinguishable ecosystems.

The concept of a critical load enables quantification of the acidifying effect and thereby enables a clear relation between emission reduction and protection against acidification. Critical loads have been defined as the highest load that will not cause chemical changes leading to long-term harmful effects. When the deposition on an ecosystem is above the critical load, the ecosystem is said to be unprotected. Depositions below the critical load do not harm the ecosystem; the ecosystem is protected against acidification. Figure 1.6 illustrates the critical load concept for one ecosystem. The deposition on an ecosystem either exceeds the critical load function, or not. Thus an ecosystem is either unprotected or protected. This binary property of the critical load concept will turn our be very important in this report.

The critical load concept is applied in RAINS and used to develop air pollution policies. However, the concept has not been without criticism. The existence of a sharp threshold like the critical load has been questioned. Also the assumption of steady-state, no dynamics, was criticised. Countries do not all calculate critical load data in the same way and the critical loads are afflicted with large uncertainties. (Skeffington, 1999)

Using the critical load concept, European acidification or acidifying impact is expressed as the total area of unprotected ecosystems in Europe. This report adopts the same measure for acidification. Because of the binary property of the critical load concept, acidification depends in a non-continuous way on deposition (and thus on emission).
Within the Convention the Working Group of Effects (see Figure 1.3) deals with research on and monitoring of pollutant effects. The several International Cooperative Programmes (ICPs) consider effects on specific kinds of ecosystems (forests, vegetation, waters), on materials, or on human health, monitoring of the state of ecosystems, or modelling and mapping. The Coordination Center for Effects (CCE) supports the ICP modelling and mapping. CCE develops “modelling and mapping methodologies for the assessment of critical loads and exceedances on a European scale” (Hettelingh et al., 2004a).

“The CCE scientifically assesses the risks, expressed as critical thresholds, to forests, lakes and other ecosystems in Europe caused by (long-range) air pollution. This information, which is also summarized in maps of critical thresholds, is useful to understand how national air pollution abatement measures can reduce these risks to ecosystems anywhere in Europe.” (http://www.rivm.nl/cce/) With the cooperation of National Focal Centres CCE collects information of all European ecosystems in the critical load database. This critical load database contains almost 1.4 million ecosystems. The area of an ecosystem ranges from a few square meters to more than 2000 square kilometres. Ecosystems in the 2004 critical load database are defined on the 50x50km$^2$ EMEP grid. One grid cell can contain thousands of ecosystems and their critical loads (see Figure 1.7). The research described in this report uses the critical load database intensively.

With the critical load database, the integrated assessment model RAINS determines the acidifying impact resulting from SO$_2$, NO$_x$, and NH$_3$ deposition. These depositions linearly relate to national emissions via SRMs. This way RAINS realizes a direct relation between emission (reduction) and (protection against) acidification. For each proposed emission reduction RAINS can determine and quantify the environmental effect on European ecosystems. The quantification of acidifying effects was used in the LRTAP Protocol on the Further Reduction of Sulphur Emissions and the Protocol to Abate Acidification.
Eutrophication and Ground-level Ozone. The LTRAP approach evolved from “effect-related” to “effect-based” assessment. (Hettelingh et al., 2004a)

Figure 1.7: example of critical load functions related to ecosystems within one grid cell.

1.3 Use of acidification factors

The main purpose of acidification factors (AFs) is to ease the calculation of acidification resulting from an emission change. Figure 1.8 gives examples of how national emission changes ($\Delta E$) can be related to changes in acidification ($\Delta A$). The examples originate from the article of Hettelingh et al. (2005) who use the unprotected ecosystem area in Europe as a measure for acidification; the same indicator as used in this report.

It is possible to model changes in acidifying impact ($\Delta A$) resulting from changes in acidifying emission ($\Delta E$). This can be realized by using a model like Eutrend or EMEP/RAINS (SRMs of RAINS, part of the DEP module in Figure 1.4) to compute dispersion and deposition, followed by applying a critical load exceedance program to calculate the area of protected ecosystems. Using such an “exact model” leads to the results shown in Figure 1.8. However, applying the “exact model” is computation intensive. Therefore AFs are introduced. AFs linearly relate the amount of national emission change to a change of protected ecosystem area in Europe:

$$\Delta A \approx AF \cdot \Delta E$$

Equation 1.1
Life cycle impact assessment (LCIA) uses acidification factors (AFs) to determine the acidifying impact from a product of service caused by emissions of substances like SO$_2$ and NO$_x$. The same linear method is applied for other impact categories like eutrophication, ozone depletion, and climate change. Conventional LCIA expresses the acidifying impact as hydrogen release potentials. Such an approach does not include distribution, exposure, and effect; only the maximum amount of hydrogen potentially released by an emission determines the acidifying impact. Potting et al. (1998a; 1998b) and Krewitt et al. (2001) introduced AFs that are based on models including distribution, exposure, and effect. They calculated their factors for each country in Europe using the integrated assessment models RAINS (Potting) or EcoSense (Krewitt). The change in unprotected ecosystem area in Europe ($\Delta A$) per change in national emission ($\Delta E$) follows from these models. Potting et al. (1998a; 1998b) and Krewitt et al. (2001) express the acidifying impact as area of unprotected ecosystem in Europe.

In an integrated assessment model like RAINS it is convenient to use linear relations as an approximation during optimization procedures. AFs can be used in RAINS in such a way (Hettelingh, 2005). Other applications of impact factors in integrated environmental assessment (IEA) include assessing external costs of energy related activities (European Commission, 2003) and computing avoidance costs (Vermoote & de Nocker, 2003). The use of AFs in IEA is still in its infancy.
1.4 Problem setting

The ratio $\Delta A/\Delta E$ (calculated from the sample points in the $\Delta A(\Delta E)$ graphs, see Figure 1.8) defines the ideal AF for a certain country, a certain substance, and a certain emission reduction. However, it is desirable to have one general AF for a certain country and a certain substance, independent of the emission reduction. It has been assumed that the AF obtained as $\Delta A/\Delta E$ was constant for a large range of emission reductions. Potting et al (1998a; 1998b) applied a change in emission per country of -10% while Krewitt et al. (2001) used $\Delta E = +10\%$ to calculate AFs which were to be applied in a large range of emission changes.

In a recent article Hettelingh et al. (2005) observed variable behaviour of $AF = \Delta A/\Delta E$ calculated using small emission changes ($\Delta E \leq 20\%$). The assumption that $AF = \Delta A/\Delta E$ is constant for a large range of $\Delta E$ turned out to be questionable. The value of AFs varies considerably when calculated with small emission changes ($\Delta E \leq 20\%$). When large emission changes are used to determine AFs, they are almost independent of the emission change used to calculate them. Therefore, Hettelingh et al. (2005) propose to use AFs calculated with an emission reduction of 50%, $\text{AF}(50\%)$, for the approximation of $\Delta A$.

LCIA generally considers small emission changes. Therefore, study of emission changes of less than 20% and AFs to calculate the corresponding acidification seems relevant. The error in the calculated acidification caused by an emission change of for example $\Delta E=10\%$ is as small as possible when calculated with $\text{AF}(10\%)$. However, as said before, it is not desirable to use different AFs for each separate $\Delta E$. One general AF is applied to approximate $\Delta A$, independent of the value of $\Delta E$. Variations in $AF = \Delta A/\Delta E$, for small values of $\Delta E$, indicate errors in $\Delta A$ calculated using the general AF instead of the emission change specific $AF = \Delta A/\Delta E$.

Hettelingh et al. (2005) discuss whether the observed variable behaviour of $\Delta A/\Delta E$ might be due to the coarse spatial resolution of sources (countries\(^1\)) and receptors (150x150km\(^2\)) which are underlying the RAINS model. They suggest to study whether the observed variability will decrease when the spatial resolution of the model is increased. The current research project explores this hypothesis by applying a variable resolution model to calculate AFs. When the variability reduces at finer resolutions, AFs calculated using small emission changes become more alike and more like $\text{AF}(50\%)$. That would indicate that the observed variable behaviour of $\Delta A/\Delta E$ is caused by a model artefact. In that case $\text{AF}(50\%)$ is suitable as general AF for the calculation of acidification caused by a large range of emission reductions, including small emission changes.

Within LCIA and also in other disciplines like IEA, the correctness of the applied AFs is of great importance. Using AFs instead of a more extended model introduces errors in the calculated acidification. These errors will be larger when AFs are applied in a situation different from the one they were originally calculated for. Several parameters influence the value of AFs, like the base year used for the emissions, the model, the way AFs are calculated, and the version of the applied database with ecosystem information. The

\(^1\) The source receptor matrices (SRM) used in RAINS relate country level emissions to deposition on the EMEP grid. SRMs are derived from the EMEP model using emissions on grid level. This aggregation of emissions could introduce errors of 10-50\% in the computed deposition (Cocks et al., 1998).
present research focuses on errors from using AFs instead of the more extended model when different spatial situations are considered. It contributes to the determination of boundaries defining the usability of AFs.

1.5 Research objective
The overall goal of the research is to explore the influence of spatial parameters (emission and deposition resolution, distribution of emissions, and actual location of emission) on AFs. Therewith we determine the correctness of using AFs in different situations and we might explain the observed variability of AFs calculated as $\Delta A/\Delta E$ using small emission changes. Ideally AFs should be insensitive for the resolution of the used model and other (spatial) parameters.

1.6 Research questions
The research objective leads to the following research questions:
1. How influences a decrease in deposition grid-size and/or an increase in emission spatial resolution the variability of $\Delta A/\Delta E$ for small emission changes?
   The result could be a more continuous behaviour for finer resolutions, indicating that deposition and/or emission resolution caused the observed variability of the acidification factors. Then the hypothesis of Hettelingh et al. (2005) would not be rejected. AF(-50%) would indeed be suitable for small emission changes as well.
2. The distribution of emissions over a country differs for each economic sector, like agriculture, transport, or production processes. Are the various sectoral AFs equal? And to what extent do they correspond to national AFs?
   The result can indicate whether or not it is allowed to use national acidification factors to calculate the effect of emission changes occurring in just one economic sector. Or, in other words, it tells us about the importance of spatial distribution of emissions on AFs.
3. How do AFs resulting from emissions from just one grid cell in a country relate to the national AF?
   The result can indicate the error made when using national acidification factors to calculate the effect of emission changes occurring in just one location (grid cell). The location of the emission will probably influence the AF.

1.7 The aggregation problem
The described problem of the influence of different spatial resolutions relates to the more general aggregation problem. This section gives a short introduction into the aggregation problem and the relation to (integrated assessment) modelling.

1.7.1 Nearly decomposable systems, hierarchy, and the aggregation problem
Complex systems can often be divided into more or less independent subsystems. Such systems are called nearly decomposable. Simon (1978) suggests that almost all large natural and social systems show this property. These systems are hierarchically structured, with hierarchy as “a boxes-within-boxes arrangement of subsystems and sub-subsystems” (Simon, 1999). Components within one subsystem interact more frequently and more intense than components from different subsystems. This property appears in all levels of the hierarchy.
Within (complex) systems theory nearly decomposable systems can be expressed as a matrix with strong interactions on and around the diagonal and weaker interaction further away. High frequency eigenvalues of these systems correspond to separate subsystems while the low frequency eigenvalues relate to the interactions between the subsystems. “The lower frequency eigenvalues can be computed on the basis of only aggregated descriptions of the subsystems, and without detailed knowledge of the within-subsystem coefficients” (Simon, 1978). Simon describes this mathematical property by assuming a system consisting of several layers of subsystems. When this system is disturbed the lowest level of subsystems are the first to reach internal steady states. Thereafter the level above this lowest level comes to steady states. The average behaviour of the subsystems (their eigenvalues) describes the system. The eigenvalues of the different levels of subsystems within the nearly decomposable system represent the dynamic behaviour of the system at different temporal ranges. Lowest level subsystems determine the very short-term behaviour. Higher levels relate to longer times. The top level defines the long-term dynamic behaviour of the system. Selecting one level of subsystems allows the study of the frequency domain of interest. It is possible to represent the details of a level subsystems with a few aggregated parameters, when studying the next level above. Only the steady state behaviour of lower subsystems is taken into account. This behaviour is replaced by some aggregated parameters. (Simon, 1999)

The idea of decomposing complex systems into almost independent subsystems relates to a supposed hierarchical structure. Giampietro (2002) defines three contiguous levels within a hierarchical system:
1. the focal level, the level that is subject of the study
2. the higher level, that defines constrains on the dynamics of the focal level
3. the lower level, that forms initiating conditions on the focal level perturbations and can be treated as noise

The influence of lower levels on the higher is known as the ‘aggregation problem’(O'Neill, 198).

Simon and Audo (1961) describe the aggregation problem as: “In exploring the aggregation problem we seek rules and criteria – exact or heuristic – that indicate what variables to aggregate and that show the circumstances under which aggregation will yield satisfactory approximations.” They list three reasons for aggregation (within economy):
1. classify all variables into smaller groups
2. study interactions within groups as though the interaction among groups did not exist
3. study the interaction among groups without regard to the interactions within each group

1.7.2 Models and aggregation
Complex natural systems can be characterised as nearly decomposable systems. This property implies the possibility of aggregation as a way to simplify the system. Models of complex systems always contain such aggregations. Although the authors use different terms, aggregation is always part of the realization of a model. The following gives a few examples.
According to Zeigler (Zeigler, 1976), modelling consists of five parts:
1. The real system
2. The experimental frame
3. The base model
4. The lumped model
5. The computer

In our situation the real system relates to the actual emissions and the ecological situation. This reality is transformed into a system by defining inputs (emissions) and outputs (ecosystem state). The experimental frame determines the limits of the validity of the model. In the base model a complete description of the real system reveals. Such a description can never be fully known because the real system is too complicated. By lumping elements of the base model a simplified lumped model remains. This step involves simplifications or aggregations. Finally the model can be implemented on a computer.

Models are simplifications of the natural world. Increasing the complexity of models (lowering the level of aggregation) does not necessarily increase the reliability. Adding more parameters and processes also adds uncertainties to the model. Thus, a more realistic (disaggregated) model does not lead to more certain predictions. Oreskes (2003) calls this effect the "complexity paradox".

Environmental models usable for management and policy should be simple. They must be easy to use, compute results fast, and require little data. To make models realistic and applicable to a broad range of boundary condition, they become complex. Many grid points, short time steps, and many processes characterize such models. Wirtz (2001) noticed that “optimal solutions satisfying both requirements of complexity and simplicity are widely missing”. Or, in other words, a general theory on optimal aggregation techniques to simplify complex systems is not available.

Muetzelfeldt and Yanai (Muetzelfeldt & Yanai, 1996) conclude the same for ecological systems: “there do not appear to be any generally accepted criteria for deciding on the level of detail included in a particular model developed to address a specific problem.” They describe three types of aggregation studies (within ecology):
1. Compare models that happen to have different aggregation levels
2. Aggregate one part of a disaggregated model
3. Theoretical analyses of certain mathematical models

The first types of studies include also other differences than just the aggregation level, making a separate study of the effects from aggregation difficult. The second type of study, which is not very often undertaken, relates to specific models and ways of aggregating making it hard to generalize the results. Finally, the third type of studies relates to only a certain kind models, again making it impossible to draw general conclusions on aggregation.

The lack of a general theory of aggregation within models had lead to a large number of publications dealing with aggregation problems for certain kinds of problems within certain scientific areas. Economic aggregation problems were recognised long ago (see for example (Klein, 1946) (Peston, 1959)). “In applied economics, basic data sets are usually derived from national accounts that contain data that are linearly aggregated over
individuals, companies, or organizations” (Constanza & Wainger, 1993). Especially aggregation related to input-output analyses have been treated extensively, a general explanation of aggregation problems in this field has been given by Ching-Han Fei (1956). In analogy to the theory developed within economy, also ecology studied aggregation issues. However, ecological problems are often defined in a non-linear way while aggregation in economy mainly concentrated on linear relations (Iwasa et al., 1987). Aggregation in ecological models is possible, but only successful if the goal of the study is narrowly defined (Bartell et al., 1988). Also within ethics (e.g. Segerberg, 1976), biological (e.g. Pinnegar et al., 2005), or sociological (e.g. Churchill, 1972) studies aggregation issues arose. Land use (e.g. Veldkamp et al., 2001), water quality (e.g. Beck, 1987), and soil (e.g. de Vries et al., 1998; McBratney, 1998) research recognized the importance of aggregation.

1.7.3 Aggregation error
“Perfect aggregation” is only possible under very strict conditions (Iwasa et al., 1987; Cale et al., 1983). Therefore, we have to settle for “approximate aggregations” (Auger et al., 2000). To quantify the success of such aggregations, an aggregation error has to be defined. Small aggregation errors indicate successful aggregation.

O’Neill and Rust (1979) presented a mathematical determination of the aggregation error in (ecological) models. They assume that the actual, real world, system consists of a large number of components. The model that represents this system contains a smaller number of parameters. At time $t$ the aggregation error follows from the difference between the state variables of the exact system and the model:

$$E(t).$$

To allow calculation of the aggregation error, O’Neill and Rust (1979) compare an exact linear system with two interacting components, with a model system consisting of only one variable. They calibrate the model by equalling the states of the exact system and the model for $t = \infty$. Ecological parameters are often derived from mass balances indicating equilibrium situations. Therefore equalling the systems for $t = \infty$ seems reasonable. Also the initial conditions (at $t = 0$) are set equal, emphasizing the transient behaviour of the system and model.

For the considered linear systems, the aggregation error is additive. Repeatedly aggregating two parameters into one, results in an addition of the corresponding aggregation errors:

$$E_{(1+2+3+4)}(t) = E_{(1+2)}(t) + E_{(3+4)}(t) + E_{((1+2)+(3+4))}(t)$$

Equation 1.2

This way the total aggregation error of a complex system can be calculated.

O’Neill and Rust (1979) minimized the integrated ($t$ from 0 to $\infty$) squared aggregation error. This error is small if the components of the exact system have similar turnover times (change with the same speed) or if one of the components is relatively rare. Therefore, minimizing the aggregation error involves lumping components with turnover times that are alike and combining rare components with common ones.
1.7.4 Spatial resolution

Three kinds of aggregation can be distinguished (see e.g. Rotmans, 1998):

- spatial resolution
- temporal resolution
- level of model complexity

The first two relate to somehow combining information on a geographical or temporal scale into a group. The last one leads to the concept of response surface methodology or meta-models (Hettelinge, 1989). In the current research we focus on the first kind of aggregation: spatial resolution.

The rise of GIS (GeoInformation System) software triggered renewed systematic attention to spatial aggregation issues. GIS eases the use of data defined at different spatial resolutions (Atkinson & Tate, 2000). Studying the influence of different spatial resolutions on the outcome of environmental models is made simpler. GIS can compare results of environmental models ran with fine or coarse aggregated input data (Mitchell et al., 2002). Using a GIS program Van Beurden and Douven (1999) showed the sensitivity of model results for aggregation before or after modelling and for the aggregation procedure (mean, threshold, worst case, or pattern indicator).

Spatial aggregation is for example studied in economics (Miller, 1998), landscape patterns (Syphard & Franklin, 2004), and groundwater recharging (Kersebaum & Wenkel, 1998). Two examples of spatial aggregation studies within atmospheric modelling deal with ozone concentration and mercury deposition: Ozone concentration on ground level has been modelled on three different scales. The spatial resolution required for quantifying the peak concentration, average concentration, or concentration at a certain time of the day differed. The finest resolution is not always required to determine these quantities (Thunis, 2001). Another atmospheric study modelled mercury deposition using three different spatial resolutions. They observed variations in the results. Since the finer resolution did not always resulted in better comparison with the measurements, it was concluded that “some key Hg chemical transformations are likely missing in current models of atmospheric Hg” (Seigneur et al., 2003).

These studies consider the influence of different spatial resolutions on some output parameter of the model. They do not include a thorough general, theoretical approach to the aggregation problem. It is recognised that aggregation problems exist and the influence is studied for specific cases. However, no generalisation in included.

Within this report we can do no more than add another example to aggregation issues in air pollution modelling. Studying different spatial resolutions relates to spatial aggregation issues, while deriving AFs relates to decreasing model complexity. The results of this study only form another example of what aggregation can do, there will (again) be no theoretical contribution to the aggregation theory.
1.8 Structure of the report

In chapter 2 we elaborate on the followed approach. Several modelling steps are introduced which lead to the creation of $\Delta A(\Delta E)$ graphs. Next, the chapter describes different analyses of these graphs using data from Hettelingh et al. (2005) as illustration. The properties we want to analyse are quantified.

Chapter 3, 4, and 5 present the main results. For each spatial aspect studied: emission/deposition resolution, sectoral emission, and grid cell specific emissions, the influence on AFs is described. This influence includes the variability of AFs calculated using small emission changes, values of AFs from different calculation methods, and corresponding errors.

During the research we discovered some other sensitivities of AFs. Acidification factors turned out to depend on, among other things, the critical load database and the numerical resolution of deposition maps. Chapter 6 shortly discusses these sensitivities.

Finally, a discussion about the results follows and the report ends with a short conclusion.
2 Approach

2.1 Boundary settings

The report considers three different spatial resolutions for the emission side:
- Emissions modelled as one source per $50 \times 50$ km$^2$ grid cell (per country/region).
- Emissions modelled as one source per $150 \times 150$ km$^2$ grid cell (per country/region).
- Emissions modelled as one source per country/region.

The last option is added to mimic the RAINS emission resolution. Although the source-receptor matrices used in RAINS were calculated with distributed emissions sources (Simpson et al., 2003, chapter 4; Vestreng, 2003) when applied in RAINS only total national emissions are distinguished.

For each emission resolution two deposition resolutions are studied:
- Deposition on $50 \times 50$ km$^2$ grid cells and
- Deposition on $150 \times 150$ km$^2$ grid cells

This leads to a total of six different emission/deposition resolution alternatives. When the report presents relative values, they are relative to the finest resolution: E50D50. (See Appendix 1 for the terminology used)

Besides spatial resolution of emission and deposition this study also considers:
- Spatial distribution of emissions (realized by studying sector specific results)
- Actual location of emissions (realized by grid cell specific results)

Both are based on the E50D50 alternative.

The influences of spatial emission/deposition resolutions are studied for the same nine country/pollutant combinations studied in the article of Hettelingh et al. (2005): SO$_2$ from Germany, France, and Italy, NO$_x$ from Belgium, Spain, and Great Britain, and NH$_3$ from the Netherlands, Poland, and Sweden. To limit the amount of computing time, all other experiments only deal with the Netherlands and France. These countries were chosen because they differ in size and because France suffers from a very local ammonia problem in the (North) Western part of the country. For these two countries the research includes all three substances, SO$_2$, NO$_x$, and NH$_3$, to incorporate substance dependent behaviour.

The study intensively uses EMEP grids, both with $50 \times 50$ km$^2$ grid cells and with $150 \times 150$ km$^2$ grid cells. Detailed explanation of the EMEP coordinates and their relation to longitude/latitude coordinates can be found in appendix A of the CCE status report of 2003 (Posch et al., 2003), or on the EMEP website (http://www.emep.int/grid/index.html).

2.2 Creation of $\Delta A(\Delta E)$ graphs

The followed research procedure exists of two main parts. First the $\Delta A(\Delta E)$ graphs are calculated and next these graphs are analysed. This section deals with the first part: the computation of $\Delta A(\Delta E)$ graphs. Figure 2.1 shows the steps required to calculate $A$ from $E$.

The following subsections describe these steps. A detailed description including file formats and examples can be found in a separate note (Bellekom, 2005). Calculating dispersion and deposition and accessing the critical load database are performed with existing software programs, all other calculations and format transformations were programmed especially for this study.
2.2.1 Emission values
Emissions originate from http://webdab.emep.int, selecting “expert emissions”. A description of these emission values can be found in Vestreng et al. (2004). We selected the year 2000 as the base year for emissions because this is also the base year of the CAFE (Clear Air For Europe) program of the European Union (Amann et al., 2004).

The EMEP website gives emission values per country/region, per 50x50km$^2$ grid cell, per substance, and per SNAP sector. Downloading these emission values from the website provides us with emission information on the finest emission resolution. Coarser emission resolutions are derived from these 50x50km$^2$ emissions. The SNAP (Selected Nomenclature for sources of Air Pollution) sector defines the economic activity where the emission originates from (see Table 2.1).

<table>
<thead>
<tr>
<th>SNAP</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>Combustion in energy and transformation industries (stationary sources)</td>
</tr>
<tr>
<td>S2</td>
<td>Non-industrial combustion plants (stationary sources)</td>
</tr>
<tr>
<td>S3</td>
<td>Combustion in manufacturing industry (stationary sources)</td>
</tr>
<tr>
<td>S4</td>
<td>Production processes (stationary sources)</td>
</tr>
<tr>
<td>S5</td>
<td>Extraction and distribution of fossil fuels and geothermal energy</td>
</tr>
<tr>
<td>S6</td>
<td>Solvent use and other product use</td>
</tr>
<tr>
<td>S7</td>
<td>Road transport</td>
</tr>
<tr>
<td>S8</td>
<td>Other mobile sources and machinery</td>
</tr>
<tr>
<td>S9</td>
<td>Waste treatment and disposal</td>
</tr>
<tr>
<td>S10</td>
<td>Agriculture</td>
</tr>
<tr>
<td>S11</td>
<td>Other sources and sinks</td>
</tr>
</tbody>
</table>

2.2.2 Emission sources
From the emission values circular emissions sources are created that serve as input for the dispersion and deposition model. For the 50x50km$^2$ and 150x150km$^2$ emission resolution the sources have an area equal to the grid cell area (2500km$^2$ and 22500km$^2$) and with their centre point at the grid cell centre. The country level emission resolution models national emissions as one circular emission source with an area equal to the country area, with the centre point at the centre of (emission) gravity.

Besides shape, location, and strength emission sources also require heat content, source height, vertical spread of source heights, and diurnal variation. These parameters differ for the various SNAP sectors. Therefore, each sectoral emission is modelled as a separate emission source.

2.2.3 Dispersion and deposition
Eutrend is an atmospheric dispersion and deposition model for the European domain that can handle various dimensions of emission sources and that supports multiple deposition resolutions. Eutrend has been selected for this study because of these reasons and because
Eutrend is NOT a grid based program. It actually determines the deposition on a given receptor point. When selecting an EMEP grid as deposition grid, Eutrend calculates the deposition on the centre point of each grid cell (van Jaarsveld, 2005). This value is taken as grid cell value. So the calculated deposition is actually the deposition at the centre point of the grid cell and is not a grid cell average.

As climatological period we selected 1990, the most recent year of which Eutrend climatological data is available. Eutrend’s output (deposition map) is defined on the 150x150km² or 50x50km² EMEP grid, both readily available within Eutrend.

**Aggregated total European deposition**

In the default situation (year 2000 emissions) the aggregated total European deposition calculated by Eutrend depends on the emission/deposition resolution (see Figure 2.2). This dependency might influence the results of our study.

Several reasons for the observed dependence of the aggregated total European deposition on the emission/deposition resolution exist. This section discusses two of them.

Considering all European emissions includes emissions at the border of the study area. A large portion of those emissions will deposit outside the study area because the emitted
substances can travel hundreds or even thousands of kilometres before depositing. A small part of the defined sources even emit outside of the study area. Especially when considering emission resolution on country level this effect occurs, since large parts of the circular sources representing national emissions exceed the study area. Circular sources of 150x150km$^2$ emit less material outside the study area and 50x50km$^2$ sources will perform best. Figure 2.3 shows the percentage of emitted substances depositing on the study area for different emission resolutions. The deposition resolution is remained equal at 50x50km$^2$. Figure 2.3 clearly shows the described behaviour for NO$_x$ and NH$_3$, for SO$_2$ hardly any dependence on the emission resolution is visible.

Another reason for the spatial resolution dependence of the aggregated deposition relates to the deposition velocity. This parameter is derived from the roughness length ($z_0$) which is location dependent. A coarse map of $z_0$ serves as input to Eutrend. The coordinate system of this map differs from the EMEP grid cells used to define the receptor grid. Combined with the fact that Eutrend calculates deposition on a point, these properties influence the actual deposition velocity used to calculate the deposition in a receptor point in an unpredictable way.

To study the influence of the here described property of Eutrend, we included averaged deposition maps. From a 50x50km$^2$ deposition map a 150x150km$^2$ deposition map is derived by averaging 9 grid cells. This new 150x150km$^2$ deposition map has the same aggregated total European deposition as the original 50x50km$^2$ deposition map. Comparing results from the averaged 150x150km$^2$ deposition map to the 150x150km$^2$ deposition map calculated directly, reveals the influence of the difference in aggregated total European deposition.

### 2.2.4 Access critical load database

Once the deposition on Europe is known, the area of unprotected ecosystems in Europe ($A$) can be determined. A critical load database contains information about the European ecosystems. This database is build by the Coordination Center for Effects$^2$ (CCE). They collect critical load data from ecosystems in all European countries. The CCE is located at the National Institute of Public Health and the Environment (RIVM) in the Netherlands, where the current study is performed.

---

$^2$ The CCE is the Data Center of the International Cooperative Programme on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects, Risks and Trends (ICP M&M) and supports the work of the Convention on Long-range Transboundary Air Pollution (LRTAP) of the United Nations Economic Commission for Europe (UNECE). (http://www.mnp.nl/cce/)
To access the critical load database we used a computer program CL. CL looks in the critical load database (we choose the latest database: 2004) to find the exact area of unprotected ecosystems per grid cell. The deposition of SO$_2$, NO$_x$, and NH$_3$ per European grid cell serves as input. For each ecosystem in a grid cell CL determines whether it is protected or not by comparing the grid cell’s deposition with the critical load function of each of the ecosystems. The areas of all (un)protected ecosystems within a grid cell add up to the (un)protected area per grid cell. Except for an explicit experiment to the influence of the CL resolution, all experiments use the 50x50km$^2$ resolution of CL.

### 2.2.5 $\Delta A(\Delta E)$ calculations

The basis of all further analyses is the $\Delta A(\Delta E)$ graph, per country and per substance. Figure 2.4 shows the steps required to calculate $\Delta A$ versus $\Delta E$. For each new European emission situation, for example 3% NH$_3$ emission reduction in the Netherlands, the procedure of Figure 2.1 produces $A$. In the default situation all emissions are like they were in the year 2000. These default emissions result in a default European deposition map with a total area of unprotected ecosystems in Europe equal to $A_0$. Changing one of the emissions (in one country, one substance) with an amount of $\Delta E$ produces $A_{\Delta E=3\%\text{-}NH_3\text{-}NL}$. The corresponding $\Delta A$ equals $A_0 - A_{\Delta E=3\%\text{-}NH_3\text{-}NL}$. This procedure is repeated for $-50\% \leq \Delta E \leq +20\%$ with steps of 1%. For each acidifying substance and for each country the $\Delta A(\Delta E)$ graph (similar to the ones in Figure 1.8) is calculated.

![Figure 2.4: schematics of the determination of $\Delta A(\Delta E)$. In the left part of the figure the steps defined in Figure 2.1 are repeated 71 times for $-50\% \leq \Delta E \leq +20\%$ with steps of 1%.

Because repeating the chain of steps given in Figure 2.1 requires a lot of computing time, a different approach was followed which uses the linearity of Eutrend. European deposition maps from national emissions (one country, one substance) are multiplied by the relative emission change to determine the deposition change. The map of changed depositions is subtracted from or added to the default European deposition map. The new deposition map is inputted into the critical load program to determine $A$. The results of this approach are exactly equal to the previously described procedure.

### 2.3 Analysis of $\Delta A(\Delta E)$

The steps described so far (Figure 2.1 and Figure 2.4) lead to the basic graphs of our analysis: $\Delta A(\Delta E)$. Figure 1.8 introduced these graphs. In this research the $\Delta A(\Delta E)$ graphs are based on:
1. Different emission and deposition resolutions ($\Delta E$ per country, per substance)
2. Sector specific emission changes ($\Delta E$ per SNAP sector in a country, per substance)
3. Grid cell specific emission changes ($\Delta E$ per grid cell, per substance)

In the analysis we extract from each of these $\Delta A(\Delta E)$ graphs:
1. The variability of AF
2. The value of the acidification factor
3. The linearization error

The definition of these quantities follows below. Data from Hettelingh et al. (2005) serve as illustration.

### 2.3.1 Variability of AF

In this report we refer to the calculation method of Hettelingh et al. (2005), Potting et al (1998a; 1998b), and Krewitt et al. (2001) as Single Value (SV) method. The SV method uses, besides the origin, one single data point of the $\Delta A(\Delta E)$ graph to calculate $AF$: $\Delta E=x \%$, $\Delta A(x)$. $AF$ therefore depends on $x^3$:

$$AF(x) = \frac{\Delta A(x)}{x}.$$  

Hettelingh et al. (2005) observed unstable behaviour of $AF(x)$ when $-20\% \leq x \leq +20\%$. This section proposes a measure to quantify this behaviour.

---

**Figure 2.5**: reproduction of figure 1 of Hettelingh et al. (2005) but only considering $-50\% \leq x \leq +20\%$ and with adapted vertical scales. ($1\text{kt}=10^6\text{kg}$)

Varying values of $AF(x)$ for $-20\% \leq x \leq +20\%$ (see Figure 2.5) inspired the current research. A quantitative measure is required to study the dependence of this variability on emission change.

---

In this context $x$ stands for an emission change. Although we work with relative emission changes, expressed as percentage of national emissions relative to 2000, $x$ represents an amount of emission change in mass units.
and/or deposition resolution. Several options exist, in this report we will use the sample variance $s^2$ and the related coefficient of variation $CV$. The $CV$ instead of the more common standard deviation has been used because we wanted a relative measure of the variability instead of an absolute one, to be able to compare the variations for different countries and substances. The sample variance $s^2$ and coefficient of variation $CV$ are defined as:

$$s^2 = \frac{\sum_{i=1}^{n} (AF(x_i) - \overline{AF})^2}{n - 1}$$

$$CV = \frac{s}{AF}$$

Equation 2.1

We are interested in the difference between $AF(x)$ and $AF(-50\%)$. Therefore, instead of $AF$ the deviation from $AF(-50\%)$ is considered in the newly defined sample variance and coefficient of variation:

$$s^2 = \frac{\sum_{i=1}^{n} (AF(x_i) - AF(-50\%))^2}{n - 1}$$

$$CV = \frac{s}{AF(-50\%)}$$

Equation 2.2

To get some feeling for the $CV$ Table 2.2 lists the values for the examples given in the paper of Hettelingh et al. (2005). The large $CV$ for SO$_2$ emissions from Italy attracts attention. A few extreme values of $AF(x)$ combined with a relatively small value of $AF(-50\%)$ (see Figure 2.5) cause the large value of $CV$. Visual inspection of the variations in Figure 2.5 suggest that NO$_x$ from Spain and NH$_3$ from Sweden relate to large variability. These examples indeed show a large $CV$ in Table 2.2. Therefore, the $CV$ seems to give a suitable quantification of the observed variability.

Table 2.2: coefficients of variation of the examples from Hettelingh et al. (2005).

<table>
<thead>
<tr>
<th>Country</th>
<th>$CV$ (relative to $AF(-50%)$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DE SO2</td>
<td>21%</td>
</tr>
<tr>
<td>BE NOx</td>
<td>20%</td>
</tr>
<tr>
<td>NL NH3</td>
<td>26%</td>
</tr>
<tr>
<td>FR SO2</td>
<td>26%</td>
</tr>
<tr>
<td>ES NOx</td>
<td>55%</td>
</tr>
<tr>
<td>PL NH3</td>
<td>32%</td>
</tr>
<tr>
<td>IT SO2</td>
<td>143%</td>
</tr>
<tr>
<td>GB NOx</td>
<td>24%</td>
</tr>
<tr>
<td>SE NH3</td>
<td>53%</td>
</tr>
</tbody>
</table>

2.3.2 Value of the acidification factor

From the $\Delta A(\Delta E)$ graphs follows also a value of the AF. Hettelingh et al. (2005) suggest to select $AF(-50\%)$, i.e. the value of $\Delta A/\Delta E$ for $\Delta E =-50\%$. However, this method relies on the value of one single sample point of the $\Delta A(\Delta E)$ graph, causing a large dependence on
the accidental value of this sample point. Including more sample points of the $\Delta A(\Delta E)$ graph in the calculation of AF averages the influences of each sample point, thereby reducing the influence of a possible extreme value. This property implies a low sensitivity of such AFs to variations in the $\Delta A(\Delta E)$ graph. Using regression to calculate AFs includes all points in the considered range. Another reason to choose regression is the fact that this method results in AFs with the lowest linearization error (standard error).

Using two different methods to calculate AFs also enables detection of dependence of the results on the calculation method.

Regression
When multiple samples from measurements or model calculations are available, regression can be used to find the “best straight line” through these points. Regression defines this “best straight line” as the line with the lowest sum of squared errors. This technique uses all sample points within the considered range, not just one single value as does the previously described SV method (see section 2.3.1).

We force the regression line through the origin, since it seems intuitively wrong to have a change in unprotected ecosystem area while the emissions do not change. At zero emission change it is to be expected that no changes occur in the area of unprotected ecosystems. The existence of a constant term would point to a wrong assessment of the air pollution policy “system”. Forcing the regression line through the origin increases the importance of the origin relative to the other sample points of $\Delta A(\Delta E)$.

While AFs calculated with the SV method depend on the value of $x$, AFs calculated with regression depend on the range of emission changes considered. Both LCA and IEA apply AFs. If they want to use the same AF, this factor should be suitable for a large range of emission changes. Generally IEA deals with larger emission changes than LCA. In this report we distinguish three ranges, roughly representing ranges of emission change interesting for IEA and LCA together, IEA alone, and LCA alone. The ranges serve illustrative purposes only and do not define absolute IEA and LCA ranges.

1. $-50\% \leq \Delta E \leq +20\%$
2. $-50\% \leq \Delta E \leq -20\%$
3. $-20\% \leq \Delta E \leq +20\%$

The corresponding AFs will be called $AF1$, $AF2$, and $AF3$. Table 2.3 compares regression AFs calculated using the three different regression ranges with AFs calculated using the SV method with $x$ equal to -50%. Figure 2.6 depicts the deviation of $AF1$, $AF2$, and $AF3$ with respect to $AF(-50\%)$. For most examples $AF3$ deviates most from $AF(-50\%)$. This can be explained by the large distance between the SV point ($-50\%, \Delta A(-50\%)$) and the range of emission changes related to $AF3$ ($-20\% \leq \Delta E \leq +20\%$). Taking into account the shapes of the $\Delta A(\Delta E)$ graphs in Figure 1.8 also explains differences between the factors. Almost linear and continuous $\Delta A(\Delta E)$ graphs, like the one for SO$_2$ emissions from France (top middle graph of Figure 1.8), result in small differences between $AF(-50\%)$, $AF1$, $AF2$, and $AF3$. On the other hand, highly non-linear and discontinuous $\Delta A(\Delta E)$ graphs, like NH$_3$ from Sweden (bottom right graph of Figure 1.8), give larger differences in AFs. Also the behaviour of the $\Delta A(\Delta E)$ graph close to the SV point ($-50\%, \Delta A(-50\%)$) is of importance. For example looking at NO$_x$ emissions from Great Britain, we see that the $\Delta A(\Delta E)$ graph...
around (-50%, \( \Delta A(-50\%) \)) has a slope different from the rest of the graph. This effect results in relative large differences between \( AF(-50\%) \), \( AF1 \), \( AF2 \), and \( AF3 \).

Table 2.3: Acidification factors calculated using regression techniques on different ranges, compared to the acidification factors calculated by the SV method using \( x=b_{50\%} \).

<table>
<thead>
<tr>
<th>[m^2/kg]</th>
<th>( AF(-50%) )</th>
<th>( AF1 )</th>
<th>( AF2 )</th>
<th>( AF3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>DE SO2</td>
<td>37.22</td>
<td>39.59</td>
<td>38.98</td>
<td>43.87</td>
</tr>
<tr>
<td>BE NO2</td>
<td>14.89</td>
<td>14.48</td>
<td>14.74</td>
<td>12.56</td>
</tr>
<tr>
<td>NL NH3</td>
<td>60.46</td>
<td>58.71</td>
<td>60.11</td>
<td>49.06</td>
</tr>
<tr>
<td>FR SO2</td>
<td>14.89</td>
<td>15.58</td>
<td>15.57</td>
<td>15.51</td>
</tr>
<tr>
<td>ES NO2</td>
<td>1.57</td>
<td>1.46</td>
<td>1.46</td>
<td>1.40</td>
</tr>
<tr>
<td>PL NH3</td>
<td>24.52</td>
<td>24.06</td>
<td>23.61</td>
<td>27.24</td>
</tr>
<tr>
<td>IT SO2</td>
<td>2.97</td>
<td>3.03</td>
<td>2.95</td>
<td>3.61</td>
</tr>
<tr>
<td>GB NO2</td>
<td>8.84</td>
<td>7.90</td>
<td>7.80</td>
<td>8.61</td>
</tr>
<tr>
<td>SE NH3</td>
<td>109.36</td>
<td>99.41</td>
<td>103.55</td>
<td>66.68</td>
</tr>
</tbody>
</table>

Figure 2.6: Difference between AFs calculated using regression (\( AF1 \), \( AF2 \), and \( AF3 \)) and \( AF(-50\%) \) for the nine examples of Hettelingh et al. (2005).

Regression techniques include all data points available while the SV method only uses one data point (-50%, \( \Delta A(-50\%) \)). The single data point required for the SV method can be an outlier in the \( \Delta A(\Delta E) \) graph. Including all available data points in the linearization, reduces the dependence of the resulting AF of the inaccuracy of one single point. We therefore expect AFs calculated using regression to be less sensitive for small variation in the \( \Delta A(\Delta E) \) graph.

Both \( AF(-50\%) \) and \( AF1 \) are supposed to cover the whole range of emission changes. Comparing the sensitivity of both factors for emission/deposition resolution, sector specific emission changes, and location specific emission changes gives insight in the influence of the calculation method on this sensitivity. The argumentation given in the previous paragraph leads to the prediction that AFs calculated using regression will be less sensitive for variation in (spatial) parameters of the model.

Comparing \( AF1 \), \( AF2 \), and \( AF3 \) values for the same model parameters can reveal preference for two emission range specific factors (\( AF2 \) and \( AF3 \)) or one overall factor (\( AF1 \)). However, the dependence on model parameters is not the only property that determines this preference. The linearization error should be small as well.
2.3.3 Linearization error

The use of linearized relations introduces an error in the calculated acidification since the original function $\Delta A(\Delta E)$ is non-linear and discontinuous. The non-linearity is caused by:

1. The relation between emission and deposition can be modelled non-linear, for example in the EMEP model (Bartnicki, 1999). However, the model we will use to calculate deposition from emission (Eutrend) is a linear model.
2. The relation between deposition and area of unprotected ecosystem is non-linear. A change in emission causes a change in deposition which can cause a certain ecosystem to change its state (protected/unprotected). The $\Delta A(\Delta E)$ function then jumps with $\Delta A$ equal to the ecosystem area, leading to discontinuous behaviour. Both the non-linearity and the discontinuity of the $\Delta A(\Delta E)$ function influence the correctness of the linear approximation of this function.

The linearization error is a measure of the suitability of an AF for the calculation of acidification. The sample error, $u$, is defined as the difference between the acidification determined with the “exact model”, $\Delta A(\Delta E)$, and the linear approximation applying an AF:

$$u(\Delta E) = \Delta A(\Delta E) - AF \cdot \Delta E$$  \hspace{2cm} \text{Equation 2.3}$$

For each sample $i$, this error can be calculated:

$$u(\Delta E_i) = \Delta A(\Delta E_i) - AF \cdot \Delta E_i$$  \hspace{2cm} \text{Equation 2.4}$$

Figure 2.7 shows an example of the sample errors $u$ when considering NO$_x$ from Great Britain (GB). The example compares the values of $\Delta A(\Delta E)$ as calculated by the “exact model” with those approximated by the linear equation $AF(-50\%) \cdot \Delta E$ or $AF1 \cdot \Delta E$. Since the SV method results in a linearization that crosses points (-50%, $\Delta A(-50\%)$) and (0,0) the error $u(\Delta E)$ for $\Delta E_i=-50\%$ and for $\Delta E_i=0\%$ equals zero. The applied regression technique forces the linearization through the origin as well, therefore also this technique results in a zero error for $\Delta E=0\%$. Appendix 2 gives the sample error for all nine examples, for four different AFs.

![Figure 2.7: example of absolute sample errors |$u$| (left) and relative errors (right), from using linearizations of $\Delta A(\Delta E)$ applying the SV method $AF(-50\%)$ or regression $AF1$.](image-url)
The right graph of Figure 2.7 displays a relative value of the sample error, defined as: $u(\Delta E_i)/\Delta A(\Delta E_i)$. Small emission changes relate to large relative sample errors because $\Delta A(\Delta E_i)$ can be rather small (small denominator gives large relative errors).

However, it is more convenient to have a general error: one measure indicating the error within a range of emission values. A general error indicates the correctness of the acidification calculated with AFs. Hettelingh et al. (2005) use the average absolute error as general error:

$$U = \frac{1}{n} \sum_{i=1}^{n} || u(\Delta E_i) ||$$

Equation 2.5

$n$ represents the number of samples.

Regression techniques as applied for the calculation of $AF_1$, $AF_2$, and $AF_3$ minimize the sum of squared errors: least square (LS) method. The standard error is an error measure based on this sum of squared errors and is therefore minimized when the regression method is applied. It should be possible to calculate an AF that minimizes the average absolute error, but such a technique is not applied in this report because it is unknown to the author. The standard error is given as:

$$\hat{\sigma} = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} u(\Delta E_i)^2}$$

Equation 2.6

An alternative way of performing a regression is using the total least square (TLS) method. This method does not consider “vertical” but “orthogonal” sample errors (see Figure 2.8). Because the vertical units, $[km^2]$, differ from the horizontal units, $[kg]$ or $[\%]$, minimizing the orthogonal error, $\sqrt{[kg]^2 + [km^2]^2}$, is not obvious. LS errors have the same unit as acidification and the goal of the linearization is to minimize the error in the calculated acidification. Therefore we selected the LS regression method.

![Figure 2.8: different sample error definitions related to total least squares (TLS) method (grey double arrow) and least squares (LS) method (black double arrow). The circle represents a sample point while the straight line represents the linearization.](image-url)
**Error range**

$U$ and $\hat{\sigma}$ depend on the considered range of emission changes. The same three emission ranges as used for regression calculations are also considered for error calculations:

1. $-50\% \leq \Delta E \leq +20\%$
2. $-50\% \leq \Delta E \leq -20\%$
3. $-20\% \leq \Delta E \leq +20\%$

The example of Figure 2.7 results in errors in a way depicted in Figure 2.9. For all nine examples similar graphs are included in Appendix 2. Figure 2.7 shows some features seen in most examples:

- Standard error calculations result in larger values than average absolute errors.
- Applying $AF3$ in a range of emission changes it is not designed for (whole range or $-50\% \leq \Delta E \leq -20\%$) results in large errors.
- The smallest standard error for an error range of $-50\% \leq \Delta E \leq +20\%$ corresponds to $AF1$, for an error range of $-50\% \leq \Delta E \leq -20\%$ to $AF2$, and for an error range of $-20\% \leq \Delta E \leq +20\%$ to $AF3$. This is inherent to the regression method.

![Graphs showing standard errors and average absolute errors U for each of the four acidification factors for each of the three error ranges.](image)

**Figure 2.9:** Example of standard errors (left) and average absolute errors $U$ (right) for each of the four acidification factors for each of the three error ranges.

Although it is interesting to study the error from using an AF on a “wrong” range of emissions, this is not the goal of this report. Therefore, in the remainder of this report we will consider only error ranges that correspond to the regression range. Table 2.4 marks the combinations that will be studied.

<table>
<thead>
<tr>
<th>error range</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-50% \leq \Delta E \leq +20%$</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>$-50% \leq \Delta E \leq -20%$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$-20% \leq \Delta E \leq +20%$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Table 2.4:** Considered error ranges for each of the four acidification factors.

Summarizing, the analysis of the $\Delta A(\Delta E)$ graphs include the fields marked in the array of Table 2.5. The following chapters present results ordered according to the columns in this array. An extra chapter (chapter 6) describes the analyses of some other (non-spatial) parameters.
Table 2.5: overview of spatial parameters and analyses included in this study.

<table>
<thead>
<tr>
<th>Analyses:</th>
<th>spatial parameters:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>emission and deposition resolution [Chapter 3]</td>
</tr>
<tr>
<td>$\Delta A(\Delta E)$ graphs</td>
<td>x</td>
</tr>
<tr>
<td>variability of AF expressed as $CV$ [%]</td>
<td>x</td>
</tr>
<tr>
<td>values of $AF(-50%)$, $AF1$, $AF2$, and $AF3$ [m$^2$/kg] (including preference of AF calculation method and emission range specific/overall AFs)</td>
<td>x</td>
</tr>
<tr>
<td>linearization error of $AF(-50%)$, $AF1$, $AF2$, and $AF3$ [km$^2$] ($U$ and $\hat{U}$) (including preference of AF calculation method and emission range specific/overall AFs)</td>
<td>x</td>
</tr>
</tbody>
</table>
3 Results from spatial emission/deposition resolution analysis

The first model parameter to be studied is the spatial resolution of emissions and depositions. Emission resolutions on country level, $150 \times 150 \text{km}^2$, and $50 \times 50 \text{km}^2$ will be combined with deposition resolutions of $150 \times 150 \text{km}^2$ and $50 \times 50 \text{km}^2$. The results indicate the sensitivity of AFs for the spatial emission and deposition resolution in the used model. Also the hypothesis from Hettelingh et al. (2005) that the variability of $AF(x)$ for small $x$ will decrease at increasingly fine resolutions of emission and deposition can be tested with the results in this chapter.

It is expected that ecosystems in the vicinity of an emission source are most likely to become protected when the emission is reduced. The majority of $\Delta A$ will probably occur close the country that decreases its emissions. In that region the largest deposition decline take place. Only ecosystems that are slightly exceeding their critical load will change their state from unprotected to protected because of an emission reduction in a neighbouring country. To check this expectation section 3.1.1 determines the location of ecosystems changing their state as a result of emission reductions.

Different emission/deposition resolutions result in different default European areas of unprotected ecosystem, $A_0$ (see Figure 3.1). These differences are partly caused by the resolution dependence of the aggregated total deposition on Europe (Figure 2.2). Ideally the difference in $A_0$ would originate only from different distributions of the deposition, not from different aggregated total depositions.

![Figure 3.1: Default European area of unprotected ecosystem, $A_0$. Left depicts absolute values; right shows values relative to E50D50.](image)

3.1 $\Delta A(\Delta E)$ graphs

For each of the nine sample situations the $\Delta A(\Delta E)$ graphs follows from the experiments. These graphs form the basis of further analyses. Ideally the sample points in the graph create a straight line. However, in practice that is not the case. The graphs show non-linear and discontinuous behaviour. Figure 3.2 shows one example of the set of $\Delta A(\Delta E)$ graphs, at different emission and deposition resolutions. The top row depicts results from a deposition resolution of $150 \times 150 \text{km}^2$ while the bottom row relates to a deposition resolution of $50 \times 50 \text{km}^2$. From left to right the emission resolution refines from country level via $150 \times 150 \text{km}^2$ to $50 \times 50 \text{km}^2$. The bottom right graph represents the finest resolution used in this study: both emission and deposition are defined on $50 \times 50 \text{km}^2$. 
It seems that the sample points form a straight line for a limited emission range followed by a “jump”. Considering for example an emission resolution of 50x50km$^2$ and a deposition resolution of 150x150km$^2$ (top right graph of Figure 3.2) the sample points seem to lie on a straight line for: $-50\% \leq \Delta E \leq -41\%$, $-40\% \leq \Delta E \leq -11\%$, $-10\% \leq \Delta E \leq +13\%$, and $+14\% \leq \Delta E \leq +20\%$. The “jumps” cause the linearity on the whole emission range (the overall linearity) to be lower than the linearity on a part of the emission range.

![Figure 3.2: $\Delta A(\Delta E)$ graphs for NO$\textsubscript{x}$ emissions from Belgium, at different emission and deposition resolutions. From left to right: emission resolutions on country level, 150x150km$^2$, and 50x50km$^2$. From top to bottom: deposition resolution of 150x150km$^2$ and 50x50km$^2$.](image)

From the example in Figure 3.2 becomes clear that a finer emission or deposition resolution does not improve the overall linearity nor does it reduce the discontinuous behaviour of the $\Delta A(\Delta E)$ graph. The bottom right $\Delta A(\Delta E)$ graph corresponding to the finest resolution of both emission and deposition does not show smaller “jumps” than the other graphs. Looking at the $\Delta A(\Delta E)$ graphs one might suggest that the discontinuous behaviour, the “jumps”, causes the overall non-linearity. Since parts of the graphs, where no large “jumps” occur, seem to show a linear relation between $\Delta A$ and $\Delta E$. The other examples are given in Appendix 3. They show similar behaviour: no improvements at finer emission or deposition resolutions.

### 3.1.1 Location of ecosystems changing their state

To analyse at what location the ecosystems change from protected to unprotected, Figure 3.3 depicts the grid cells that change when the emission of the nine sample country/substance combinations is reduced by 50%. The figure results from three effects:

1. Each of the substances has a different range (transport distance), NH$_3$ deposits closer to the source than NO$\textsubscript{x}$.
2. The dominant wind direction, from south-west to north-east, determines in which direction from the source the substances deposit.
3. Finally, the sensitivity and extent of exceedance of the receiving location makes the ecosystem to flip from unprotected to protected, or not.

Figure 3.3 indicates that emissions can cause effects at a long distance, even if the deposition at such distances has become rather small. For example, NH$_3$ emissions from the Netherlands cause effects as far away as Poland while the majority of the emitted NH$_3$ deposits close to the emission source. Most effects occur in the Northern part of Europe.
which can be explained by the more sensitive ecosystems in that part of Europe. Those ecosystems are also more often unprotected in the default (year 2000) situation and can become protected by a decrease in deposition (resulting from a decreasing emission).

Figure 3.3: grid cells in which the area of unprotected ecosystem changes when the national emission is reduced with 50%.
From Figure 3.3 one can conclude that taking into account only local (close to the emission source) effects can not represent the total effect. It seems necessary to include all of Europe to cover the European effect caused by changing national emissions. One can question the appropriateness of results from national emission changes of countries on the border of the considered area. A large part of the effect of emissions from those countries will take place outside of the considered area of Europe.

“Jumps” in the $\Delta A(\Delta E)$ graphs need further analysis. Although between every two successive points with a different value of $A$, a “jump” exists, we concentrate on the large visible “jumps”. Since the emission/deposition resolution E50D50 is the finest one available, finding the location of the “jumps” focuses on that resolution. Figure 3.4 depicts the nine $\Delta A(\Delta E)$ graphs and the considered “jumps”. Each of these “jumps” in $\Delta A$ relate to a 1% change in national emission. Figure 3.5 indicates where the ecosystems that change their state at these “jumps” are located. It seems that in Norway some large, sensitive ecosystems exist; changing SO$_2$ from DE, NO$_x$ from BE, and NO$_x$ from GB cause a large effect in Norway. This can be seen in the top left, middle left, and middle right maps of Figure 3.5 as a small black square in the southern part of Norway. In the top left (SO$_2$ from DE) and middle left (NO$_x$ from BE) maps this grid cell is located at the east side of southern Norway. In the middle right map (NO$_x$ from GB) the large Norwegian ecosystem is found more to the west.

Figure 3.4: $\Delta A(\Delta E)$ graphs of the nine examples, with an emission and deposition resolution of 50x50km$^2$. Ovals indicate which “jumps” are studied. The corresponding locations of these “jumps” are depicted in Figure 3.5.
The changing area in grid cell (51,60)\(^4\) in Southern Norway, caused by emission changes of SO\(_2\) from DE or NO\(_x\) from BE (see corresponding graphs of Figure 3.5), equals 200.6km\(^2\). This grid cell contains one ecosystem with an area of 200.3km\(^2\) which is the ecosystem changing its state. Figure 3.6 shows this situation. In the left graph the range of

\(^4\) EMEP 50x50 coordinates (i,j).
sulphur depositions is hardly visible because the changes are very small. The right graph enlarges the graph around the deposition range. In this graph the top of the line with deposition values (diamonds) represents the default situation (year 2000 emissions). When the SO$_2$ emission reduction in Germany is 0% the ecosystem is slightly unprotected. Reducing SO$_2$ emissions in Germany causes the sulphur deposition in grid cell (51,60) to decrease. Between 22% and 23% reduction of these emissions the considered ecosystem becomes protected.

Also the changing area in grid cell (49,60), caused by emission changes of NO$_x$ from GB, equals the area (436.3 km$^2$) of one of the ecosystems defined in that grid cell.

We conclude that large “jumps” in $\Delta A(\Delta E)$ graphs are mainly caused by one large ecosystem changing its state. The size of these large “jumps” is determined by the size of the ecosystems in the database. However, Figure 3.5 shows that a change in national emissions of 1% causes several smaller ecosystems to change their state as well. In the large visible “jumps” in the $\Delta A(\Delta E)$ graphs beside small ecosystems also one large ecosystem changes its state and that ecosystem area dominates the change in $\Delta A$. This large ecosystem does not necessarily be located close to the emission source.

The large visible “jumps” in the $\Delta A(\Delta E)$ graphs cause the linear approximation of the $\Delta A(\Delta E)$ function to have large errors. Reducing these linearization errors includes reducing the size of the “jumps”. Since the size of large visible “jumps” is mainly determined by the area of one ecosystem, refining the ecosystem database can reduce the “jumps” and improve the accuracy of acidification calculations with AFs. However, the use of AFs means that a global approximation of the acidification is strived for and such an approach should not depend on data from one single ecosystem.

Ecosystems at some distance from the emission source will only change their state when they are only slightly (un)protected in the default situation. The example of Figure 3.6 showed that small deposition changes caused by emission changes in a remote country can make such an ecosystem change its state. Ecosystems closer to the emission source receive a larger deposition change than the remote ecosystems. They can be more exceeded in the default situation and still become protected because of the emission reduction. The level of exceedance of ecosystems in the default situation influences the $\Delta A(\Delta E)$ graphs.
“Jumps” occur in all $\Delta A(\Delta E)$ graphs, not just in the E50D50 (finest emission and deposition resolution: 50x50km$^2$) case considered here but also when other emission and deposition resolutions are considered (see Appendix 3). The “jumps” seem to be independent of the used emission and/or deposition resolution.

### 3.2 Variability of AF

The first analysis of the $\Delta A(\Delta E)$ graphs relates to the variability of $AF(x)$ for small $x$. Hettelingh et al. (2005) suggested a reduction of the variability at finer model resolutions of emission and deposition. This hypothesis is tested.

Using the definition of variability as defined in Equation 2.2, the coefficient of variation of the nine examples depends on the resolution as given in Figure 3.7. The figure shows no decrease in variability when the resolution is refined. For SO$_2$ emissions from France (top middle graph), the finest emission and deposition resolution (right most bar) even shows the largest variability. Thus, the results do not support the hypothesis of Hettelingh et al. (2005).

#### Figure 3.7: variability of AFs calculated with the SV method using emission changes between -20% and +20%. The coefficient of variation (CV) relates to AF(-50%). Note that all vertical axes have very different scales.
From the figure we also conclude that the country level emission resolution does not mimic the situation of the article of Hettelingh et al. (2005) very well. Comparing the left most bars of Figure 3.7 with the values in Table 2.2 shows large discrepancies. For example, the CV of SO$_2$ emissions from France equals 26% when using data from the article, while our model gives 44% for country level emission resolution and 150x150km$^2$ deposition resolution.

![Graphs of AF(x) for NO$_x$ emissions from Belgium, at different emission and deposition resolutions.](image)

Figure 3.8: $AF(x)$ graphs for NO$_x$ emissions from Belgium, at different emission and deposition resolutions. The results correspond to the $\Delta A(\Delta E)$ graphs of Figure 3.2.

Figure 3.8 shows the graphs of $AF(x)$ for -50% ≤ $x$ ≤ +20% corresponding to the $\Delta A(\Delta E)$ graphs of Figure 3.2. The effect of “jumps” in the $\Delta A(\Delta E)$ graph on $AF(x)$ is visible. Large “jumps” occurring at large emission changes, for example in the EcD50 situation at emission reductions between 20% and 21% (bottom left graph of Figure 3.2), cause considerable variability of $AF(x)$ (bottom left graph of Figure 3.8). However, when the “jump” occurs at very small emissions changes, even a small “jump” causes a large variation in $AF(x)$. This last effect is visible when looking at the E150D50 situation (considering middle bottom graph of Figure 3.2 and Figure 3.8) where a small “jump” in $\Delta A$ between emission reductions of 1% and 2% causes $AF(x)$ at that point to change considerably. Concluding: the accidental location (of the $\Delta E$) of large “jumps” in the $\Delta A(\Delta E)$ graph determines to a large extend whether it has effect on the variability of $AF(x)$ or not.

For visual inspection of the variations, Appendix 3.2 depicts graphs of $AF(x)$ for -50% ≤ $x$ ≤ +20% for all nine examples. The graphs correspond to the $\Delta A(\Delta E)$ graphs in Appendix 3.1. Comparing these graphs leads to the same conclusions as for the example described.

---

5Differences occur because:
- Hettelingh et al. (2005) used a different dispersion and deposition model, they applied SRMs (source receptor matrices) from RAINS instead of Eutrend.
- The country to grid SRMs in RAINS do embody more detailed national emission distributions while the area sources used in Eutrend show a homogeneous distribution of the emissions.
- Circular country level area sources in Eutrend show a different shape than the country itself.
3.3 Value of AFs

The next analysis deals with the dependence of the AFs on emission and/or deposition resolution of the model. Ideally AFs do not depend on this resolution. Again, the $\Delta A(\Delta E)$ graphs (see Appendix 3.1) serve as basis for the analysis.

Figure 3.9 depicts for each of the nine examples $AF(-50\%)$, $AF1$, $AF2$, and $AF3$ versus resolution. Although the acidification factors depend on the resolution, no pattern can be found (the pattern is also not related to $A_{b}$, see Figure 3.1). To study the dependence on resolution in more detail Figure 3.10 shows for all AFs the change in value relative to the finest resolution ($E50D50$). Especially $SO_2$ emissions from Italy (top right graphs) show very large differences between AFs at $E50D50$ and at $E150D50$. This is probably caused by a large "jump" in the $\Delta A(\Delta E)$ graph of the $E150D50$ situation which is not so prominent in the $E50D50$ situation (see Appendix 3.1). On the other hand, AFs for $SO_2$ emissions from Germany (DE) only slightly depend on the spatial resolution of the model. The $\Delta A(\Delta E)$ graphs of Germany show hardly any "jumps", only $EcD50$ shows a small visible "jump" at -18%. Therefore, AFs derived from the $EcD50$ situation show the largest deviation from the $E50D50$ situation (top left graph of Figure 3.10). As in the previous analysis, large "jumps" in the $\Delta A(\Delta E)$ graphs influence the variation of AF values as a function of resolution.
Comparing dependence on resolution of $AF(-50\%)$ and $AF1$ (first two sets of bars in Figure 3.9 and Figure 3.10) gives insight in the role of the calculation method. For all nine examples the two AF calculation methods show a similar pattern of dependence on resolution. Also the absolute dependence is about equal for the two AF calculation methods. In this experiment the calculation method of the AFs does not produce distinguishing results.

To study the preference of one overall factor or two factors covering only a part of the emission range, $AF1$, $AF2$, and $AF3$ are compared. Looking at SO$_2$ from Germany $AF3$ depends less on the model spatial resolution than $AF1$ and $AF2$ (top right graph of Figure 3.10). However, for SO$_2$ from France about the opposite is true (top middle graph of Figure 3.10). Therefore, based on the sensitivity for spatial resolutions, no preference for one overall AF or two separate emission range specific AFs reveals.

### 3.4 Error

The last analysis of the $\Delta A(\Delta E)$ graphs considers the linearization error. Both the standard error $\hat{\sigma}$ and the average absolute error $U$ are included in the analysis. As explained in section 2.3.3 we consider only error ranges equal to the emission ranges used in the calculation of the corresponding AFs. Small linearization errors indicate that acidification calculated using AFs gives a good approximation of the acidification that would result from applying the complete model. Besides the magnitude of the error also the dependence on model parameters is of importance. Small errors independent of the spatial emission and deposition resolution of the model are preferred.

Figure 3.11 shows $\hat{\sigma}$ and Figure 3.12 shows $U$. For each AF the dependence of these errors on the spatial resolution is depicted. Both errors show similar dependences on the spatial resolution. It is not possible to select one spatial resolution that shows for all nine
examples the smallest error. Appendix 3.3 depicts explicitly how both errors change relative to E50D50.

NOx from Great Britain (middle right graph) shows the largest errors. The range of $\Delta A$ related to $-50\% \leq \Delta E \leq +20\%$ is also largest for this example (see Appendix 3.1). Our error definitions give results coupled to the range of $\Delta A$. A relative error would have been better.

Figure 3.11: standard error related to $AF(-50\%)$, $AF1$, $AF2$, and $AF3$ as a function of emission and deposition resolution of the model. $AF(-50\%)$ relates to the SV method using an emission reduction of 50%. $AF1$, $AF2$, and $AF3$ are derived using regression on respectively $-50\% \leq \Delta E \leq +20\%$ (whole emission range), $-50\% \leq \Delta E \leq -20\%$, and $-20\% \leq \Delta E \leq +20\%$.

Figure 3.12: mean absolute error related to $AF(-50\%)$, $AF1$, $AF2$, and $AF3$ as a function of emission and deposition resolution of the model. $AF(-50\%)$ relates to the SV method using an emission reduction of 50%. $AF1$, $AF2$, and $AF3$ are derived using regression on respectively $-50\% \leq \Delta E \leq +20\%$ (whole emission range), $-50\% \leq \Delta E \leq -20\%$, and $-20\% \leq \Delta E \leq +20\%$.  

49
Comparing errors related to AF(-50%) and AF1 (see Figure 3.11 and Figure 3.12) to extract the influence of the calculation method does not result in a preference of one or the other. For both AFs the dependence of the error on the considered spatial emission and deposition resolution of the model is similar (similar pattern of first two sets of bars in Figure 3.11 and Figure 3.12), as it was for the value of the AFs. For example consider the standard error related to NOx emissions from Spain, centre graph of Figure 3.11. Focussing on AF(-50%) one sees that the error related to EcD150 is smallest, followed by E50D150, EcD50, E50D50, E150D150, and E150D50. When studying the standard error for AF1 the same dependence of the error on the emission/deposition resolution is obtained. Thus, comparing the sensitivity of the error on the resolution does not reveal a preferred method to calculate AF.

Besides the sensitivity on the error, also the magnitudes of the errors are important when comparing AFs. The standard error is smaller for AF1 than for AF(-50%). This follows from comparing the bars in Figure 3.11 related to AF1 and AF(-50%) per emission/deposition resolution. For example the lightest grey bar (EcD150) of AF1 is always (for all nine examples) smaller than or equal to the corresponding bar of AF(-50%). Thus, when considering $\hat{\sigma}$, regression is preferred to the SV method to calculate AFs (as was theoretically expected). However, Figure 3.11 shows that the differences are small for most examples. Figure 3.12 implicates that the same is true when considering $U$, but this is not necessarily true for all examples when looking at the error values in more detail.

When comparing AF1, AF2, and AF3 the figures only show a slight preference for using two different AFs instead of one overall AF. The standard error for AF1, is in most examples larger than the standard error for AF2 or AF3. To a lower extent the same is true for $U$. Based on the magnitude of the error we therefore conclude that applying two emission range specific AFs instead of one overall AF is slightly preferred.

### 3.5 Aggregated deposition method

All previous results originate from emission and deposition resolutions applied directly to Eutrend. In Eutrend the total European deposition depends on the applied spatial resolution (see section 2.2.3). To study the influence of this effect on the results of the previous sections, this section introduces the aggregated deposition method.

The aggregated deposition method only includes deposition resolutions; the emission resolution is set at 50x50km$^2$. Based on a 50x50km$^2$ deposition map from Eutrend a new 150x150km$^2$ deposition map follows from successively averaging 9 grid cells. This way the total deposition of the 150x150km$^2$ map equals that of the 50x50km$^2$ map. This new situation is called E50D50->D150. The following subsections compare E50D50, E50D150, and E50D50->D150, the first two are copied from the previous sections but included here as reference.

Figure 3.13 shows the default area of unprotected European ecosystem, $A_0$, of the new E50D50->D150 situation (right most bar) compared to E50D50 and E50D150. Both 150x150km$^2$ deposition resolutions result in almost equal $A_0$: 189386km$^2$ and 201799km$^2$ (difference is 6%). The difference in $A_0$ when considering 50x50km$^2$ and 150x150km$^2$ deposition resolutions is larger: the $A_0$ of E50D150 is 16% lower than the $A_0$ of E50D50 and 10% lower than the $A_0$ of E50D50->D150. Therefore, the difference between E50D50 and E50D150 is not (solely) caused by Eutrend artefacts (Figure 3.1).
3.5.1 \( \Delta A(\Delta E) \) graphs

Figure 3.14 and Figure 3.15 show for two examples the E50D50 and E50D150 \( \Delta A(\Delta E) \) graph (copied from Appendix 3.1) versus the situation in which the 150x150km\(^2\) deposition map followed from aggregation of the 50x50km\(^2\) map. It seems that the last situation deviates considerable from the both the E50D50 and E50D150 situation. Comparing the left and right graphs indicates differences related to different deposition resolutions, without the confounding effect of changing aggregated total European deposition. Comparing the middle and right graphs illustrates differences between a deposition resolution of 150x150km\(^2\) resulting from direct Eutrend calculations and from averaging Eutrend results on a 50x50km\(^2\) receptor grid.

From these examples one can conclude that the E50D50->D150 situation gives different results than the E50D150 situation. This is also visible in the other examples which are included as Appendix 4. The method to create a deposition map of 150x150km\(^2\) (either as direct Eutrend output or as averaged 50x50km\(^2\) output) influences the \( \Delta A(\Delta E) \) graphs.
3.5.2 Variability of AF
The variability of $AF(x)$ calculated with small values of $x$ depends on the emission and deposition resolution, as we concluded in section 3.2. This section recalculate the dependence on the deposition resolution using the aggregated deposition method.

Comparing the $CV$ of the E50D50 and the E50D50->D150 situation (lightest and darkest bars in Figure 3.16) does not change the conclusion about Hettelingh’s hypothesis (derived in section 3.2). For some examples ($SO_2$ from DE, FR, and IT, and $NO_x$ from BE) the finer deposition resolution results in a lower variability, for the other examples the opposite is true. So, also when using the averaged 150x150km$^2$ deposition map, no systematic dependence of the variability on the deposition resolution can be concluded.

![Figure 3.16](image)

Figure 3.16: Variability of AFs calculated with SV method using emission changes between -20% and +20%.

The coefficient of variation ($CV$) relates to $AF(-50\%)$.

3.5.3 Values of AFs
Figure 3.17 shows the values of acidification factors calculated using different methods (SV method and regression) and emission ranges (regression on $-50\% \leq \Delta E \leq +20\%$, $-50\% \leq \Delta E \leq -20\%$, and $-20\% \leq \Delta E \leq +20\%$). The values of AFs vary with the deposition resolution.

Although the results for E50D50->D150 differ from the E50D150 results, the conclusion about the deposition dependence does not change. Sometimes AFs from the E50D50->D150 situation are larger than those from the E50D50 situation (e.g., all AFs related to $SO_2$ from DE: top left graph) and sometimes the opposite is found (e.g., all AFs related to $NH_3$ from PL: bottom middle graph). The AFs do not systematically depend on the deposition resolution, independent of the way the coarser deposition map is calculated.
3.5.4 Error

Considering the error from using an AF instead of the complete model, the calculation method of the 150x150km$^2$ deposition resolution does not influence the results. Independent of the way the coarser deposition resolution is calculated the error is not a systematic function of the deposition resolution. Appendix 4.2 depicts the standard error and mean average error versus the three considered deposition resolutions. From these graphs the almost arbitrary behaviour is visible.

3.6 Summary and conclusions

The analysis of the influence of emission and deposition resolution revealed that the variability of $AF(x)$ for small $x$ does not decrease at finer resolutions, not even at the finest considered resolution E50D50. This indicates that the hypothesis stated by Hettelingh et al. (2005) cannot be confirmed.

Both the AFs themselves and the corresponding error depend on the emission and deposition resolution in a non-systematic way. For example, consider the calculation of the influence of reducing SO$_2$ emission from France. When using $AF(-50\%)$ computed with an emission and deposition resolution of 150x150km$^2$, the calculated reduction in acidification will be 40% higher than when calculated using $AF(-50\%)$ computed with an emission and deposition resolution of 50x50km$^2$ (see top middle graph of Figure 3.10, third bar from the left). AFs calculated with different resolutions can differ more than 100%; E150D50 compared to E50D50 for SO$_2$ emissions from Italy shows this maximum (see top right graph of Figure 3.10). AFs strongly depend on the emission and deposition resolution used to compute them, and therefore so does the acidification that is calculated using these AFs.
Considering spatial emission and deposition resolution it is hardly possible to prefer one kind of AF above the other. A slight preference for emission range specific AFs follows from the smaller error of these factors.

“Jumps” (large discontinuities) in the $\Delta A(\Delta E)$ graphs have a large influence on AFs and errors. These “jumps” are often dominated by just one single ecosystem changing its state. That ecosystem can be located at quite some distance from the emission source. It is therefore necessary to include a large area when performing such calculations.

All previous results originate from emission and deposition resolutions applied to Eutrend. Because it is known that in Eutrend the total European deposition depends on the applied spatial resolution, the aggregated deposition method was followed to bypass this property. The analysis of the variability, value of AFs, and errors does not show a systematic improvement when calculated with the aggregated deposition method. Concluding: for an emission resolution of $50\times50\text{km}^2$ and a deposition resolution of $150\times150\text{km}^2$, Eutrend’s dependence of aggregated total deposition on the resolution does not influence the resulting AFs. This example indicates that Eutrend’s dependence of aggregated total deposition on the resolution is not the cause of the observed behaviour of AFs with respect to emission and deposition resolution. The results based on direct Eutrend calculations (section 3.1 to 3.6) seem valid.
4 Results from sector specific emission changes

After studying the influence of spatial emission and deposition resolution, this chapter focuses on sector specific emission changes. The analysis helps to determine the admissibility of using national AFs instead of sector specific AFs to calculate acidifying impacts from sectoral emissions. Since the emissions of each sector are distributed over a country in a different way, the influence of spatial distribution of emissions is studied as well. Table 4.1 repeats the definition of the SNAP sectors as used in this study.

Table 4.1: Definition of SNAP sectors (source: webdab.emep.int/sectors.html). (Copy of Table 2.1)

<table>
<thead>
<tr>
<th>Sector Code</th>
<th>Sector Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>Combustion in energy and transformation industries</td>
</tr>
<tr>
<td></td>
<td>(stationary sources)</td>
</tr>
<tr>
<td>S2</td>
<td>Non-industrial combustion plants (stationary sources)</td>
</tr>
<tr>
<td>S3</td>
<td>Combustion in manufacturing industry (stationary sources)</td>
</tr>
<tr>
<td>S4</td>
<td>Production processes (stationary sources)</td>
</tr>
<tr>
<td>S5</td>
<td>Extraction and distribution of fossil fuels and geothermal energy</td>
</tr>
<tr>
<td>S6</td>
<td>Solvent use and other product use</td>
</tr>
<tr>
<td>S7</td>
<td>Road transport</td>
</tr>
<tr>
<td>S8</td>
<td>Other mobile sources and machinery</td>
</tr>
<tr>
<td>S9</td>
<td>Waste treatment and disposal</td>
</tr>
<tr>
<td>S10</td>
<td>Agriculture</td>
</tr>
<tr>
<td>S11</td>
<td>Other sources and sinks</td>
</tr>
</tbody>
</table>

For France and the Netherlands, this chapter considers all three substances: SO\(_2\), NO\(_x\), and NH\(_3\), and the sectoral distribution. Figure 4.1 shows the distribution of the emissions over the SNAP sectors for the six examples. All examples use an emission and deposition resolution of 50x50km\(^2\).

NH\(_3\) emissions are strongly dominated by SNAP sector 10: agriculture (right graphs of Figure 4.1). Results from this sector will therefore be very much like the national results. The other substances SO\(_2\) and NO\(_x\) also show some dominant sectors, but in a less extreme way. Sector 9, waste treatment and disposal, causes only very small amounts of emissions in the Netherlands and France.

Figure 4.1: emission distribution along the different SNAP sectors for SO\(_2\), NO\(_x\), and NH\(_3\) emission from the Netherlands and France.

4.1 \(\Delta A(\Delta E)\) graphs

Like in the previous chapter, graphs of \(\Delta A(\Delta E)\) result from the calculations. For each sector and for the national emissions these graphs form the basis of further analysis. Figure 4.2 shows the result for NO\(_x\) emissions from the Netherlands as example.
Figure 4.2: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from the Netherlands. The national total (top left graph) is compared to sector specific results.

Figure 4.2 shows that the area of ecosystems in Europe changing their state (range of $\Delta A$ on the vertical axis of the graphs) becomes smaller as the corresponding Dutch emissions (see Figure 4.1) become smaller. The graph corresponding to NO$_x$ emissions within SNAP sector 9 is the most extreme one. Because of the very small emission changes involved, most emission changes do not cause any European ecosystem to change state. The deposition maps do change continuously, the steps are caused by the binary nature of the method, in which ecosystems are either protected or unprotected.

Several sectors in Figure 4.2 show a similar “jump” in $\Delta A$. For example, sector 1, 2, 3, and 8 all show a large visible “jump” of about 85 km$^2$; these “jumps” are marked in Figure 4.2. Table 4.2 further analyses these “jumps”; it gives exact $\Delta A$-steps and the related emission change. If a major part of each of these “jumps” is caused by one and the same ecosystem, then the table indicates that not only the magnitude of $\Delta E$ but also the emission distribution (sector) is important since the $\Delta E$ at which the “jump” occurs differs per sector. Theoretically the table could deal with two ecosystems, each of about 85 km$^2$. However, this seems very unlikely since none of the $\Delta A(\Delta E)$ graphs of sector 1, 2, 3, or 8 shows a second “jump” of the same size. Within the national $\Delta A(\Delta E)$ graph $\Delta A(1\%)$ equals 107km$^2$; this value includes the 85km$^2$ seen as “jumps” in the sectoral graphs. In the national graphs such values of $\Delta A$ cause no visible “jumps”. Thus, one relatively large ecosystem can cause visible “jumps” in $\Delta A(\Delta E)$ graphs of different sectors, while the same ecosystem changing its state does not cause a larger than average $\Delta A$-step in the national situation. Therefore, sectoral $\Delta A(\Delta E)$ graphs can show more “jumps” and more severe “jumps” than the national $\Delta A(\Delta E)$ graph. This effect causes sectoral AFs to be more variable than national AFs (see Figure 4.3).
Table 4.2: analysis of equal visible “jumps” in the sectoral $\Delta A(\Delta E)$ graphs of Figure 4.2. The “jumps” are marked in Figure 4.2.

| Sector 1 | 85.47 | 1.34 | 2.01 |
| Sector 2 | 83.73 | 0.54 | 0.80 |
| Sector 3 | 84.48 | 1.31 | 1.64 |
| Sector 8 | 87.04 | 0.76 | 0.97 |

Figure 4.3: coefficient of variability of $AF(x)$ for $-20% \leq \Delta E \leq +20\%$ with respect to $AF(-50\%)$. For each of the other five examples Appendix 5.1 displays the $\Delta A(\Delta E)$ graphs per sector. These examples show the same patterns. For NH$_3$, step-wise $\Delta A(\Delta E)$ graphs are observed often for sectors other than agriculture (sector 10), because of the very small emission changes involved. Figure 4.3 depicts the variability of sectoral AFs that result from the $\Delta A(\Delta E)$ graphs in Appendix 5.1. For most examples sectoral AFs are more variable than national AFs, like the described example: NO$_x$ from the Netherlands.

4.2 Value of AFs

When considering emission reduction within one economic sector, it is questionable if the national AF gives a good approximation of the reduction in acidifying impact. The goal of this section analysis is to quantify the difference of sectoral AFs with respect to the national AF. The difference between the national AF and sector specific AFs is a measure for the error made when using national factors instead of sector specific factors. Since sector specific emissions are also distinguishably distributed, the sensitivity of AF for emission distribution patterns results from this analysis as well.

Figure 4.4 depicts the values of acidification factors for national and sectoral emissions. Not only the factors calculated using the SV method, but also acidification factors resulting from regression techniques are included. The figure shows that for NH$_3$ the sectoral AF of sector 10 about equals the national AF. This can be explained by the fact that almost all NH$_3$ emissions take place in sector 10 (agriculture), see Figure 4.1.

The top middle graph of Figure 4.4 shows the AFs of the example of Figure 4.2. Remarkable is the large national value for $AF(-50\%)$. Looking at the top left graph of
Figure 4.2 one can see that the two values ∆A(-49%) and ∆A(-50%) deviate from the trend in ∆A(∆E). The “jump” in the national ∆A(∆E) graph occurring between ∆E=-48% and ∆E=-49% is not seen in any of the sectoral ∆A(∆E) graphs. Dutch NOx emission reductions between 48% and 49% relate to 203Gg and 207Gg. Such large emission reduction are not considered for the sectoral emissions; the maximum reduction in this analysis is 50% which relates for the considered example to 0.1Gg (sector 9) to 96Gg (sector 7). The described effect causes AF(-50%) to vary more across sectors than the AFs based on regression, for NOx emissions from the Netherlands.

Figure 4.4: values of national and sector specific acidification factors.

To emphasize the differences between sector specific and national acidification factors Figure 4.5 shows this difference explicitly. Comparing the NH3 from the Netherlands example of Figure 4.5 (top right graph) with the corresponding example of Figure 3.10 (bottom left graph) shows that this example is more sensitive for emission distribution (sectors) than for emission and deposition spatial resolution. The opposite is true when considering the other example: SO2 emissions from France. It is therefore not possible to conclude which of the two sensitivities is strongest.

Figure 4.5: deviation from national AFs of sector specific AFs (derived from Figure 4.4).

The influence of the calculation method of the AFs is represented by different result for AF(-50%) and AF1. Considering the dependence of these factors on the sector specific emissions (Figure 4.5) reveals no absolute differences. For NOx and NH3 from the Netherlands and for NOx from France, AF1 shows less sensitivity for all sectors than AF(-50%). In the other examples this is not true for all sectors, e.g. for sector 2, 7, and 9.
related to SO₂ from the Netherlands. AF₁ shows a larger deviation from the national factor than AF(-50%). Considering all sectors in each of the six examples, AFs calculated using regression are in general less dependent on the sector than AFs calculated with the SV method. This observation supports the theoretical prediction given in section 2.3.2.

Whether to prefer one overall AF or two emission range specific AFs follows from comparing AF₁ with AF₂ and AF₃. Some examples (SO₂ from NL and NH₃ from NL) show very large deviations from the national AF for AF₃. Large visible “jumps” in the ∆A(∆E) graph within the range of emission changes related to AF₃ (-20% ≤ ∆E ≤+20%) have a stronger effect than equally large “jumps” at larger emission reductions. All sectoral ∆A(∆E) graphs of SO₂ from NL (see Appendix 5) show a large visible “jump” in that range. Also a lot of the other sectoral ∆A(∆E) graphs include a “jump” in the range of small emission changes. As explained in section 4.1 the same “jump” can appear in several sectoral ∆A(∆E) graphs, affecting all sectoral AFs. However, there is no reason known as yet why these “jumps” should occur more often in the small emission range. The fact that for the two sample countries such a “jump” occurs in this range can be a coincidence. If that is the case, then the higher sensitivity of AF₃ for sectoral difference can also be a coincidence.

4.3 Error

Not only the dependence of the values of the AFs on the sector but also the dependence of the corresponding errors on the sectors is of importance when determining the suitability of national AFs for calculating impact from sectoral emission changes. However, this analysis showed the unsuitability of the defined standard error and mean absolute error for this purpose. These two errors very much relate to the range of ∆A and ∆E. As for example Figure 4.2 shows: for sectoral emission changes between -50% and +20% the change in A can be much smaller than the national one. Both the standard error and the mean absolute error are by far largest for the national AF. Appendix 5.2 lists the standard errors and mean absolute errors for sector specific acidification factors. The described effect is clearly visible.

However, in this chapter we are mainly interested in the error that occurs when national acidification factors are used instead of sector specific factors. To quantify this error we use two different approaches. The first one uses Equation 2.5 and Equation 2.6; the second approach applies statistics to the AFs themselves.

The first approach combines the linearization error with the error from using national instead of sectoral AFs. A new sample error is defined as:

\[ u(\Delta E_i) = \Delta A(\Delta E_i) - AF_n \cdot \Delta E_i \]

Equation 4.1

In contrast to Equation 2.4 the sample error \( u(\Delta E_i) \) of Equation 4.1 is calculated relative to the linearization using the national AF: \( AF_n \), while \( \Delta E_i \) relates to a sectoral emission change. Using the definition of Equation 4.1 in Equation 2.5 and Equation 2.6 gives an average absolute error and a standard error related to using national AFs for sector specific applications. Figure 4.6 shows the results of this exercise for the standard error. The heights of the bars represent the error in the calculated sector specific acidification when using the national AF to compute ∆A instead of the complete model. The average absolute error gives similar results. Again, it is clearly visible that these errors strongly relate the
magnitude of the sectoral emission. For example, the graph of NO\textsubscript{x} from NL (Figure 4.6 top middle) shows largest errors for sector 7 and sector 8, Figure 4.1 depicts that these sectors also emit the largest amounts of NO\textsubscript{x}.

A second approach calculates the weighted standard deviation of sector specific acidification factors. The weighting factors are defined by the emission amounts per sector. This approach leads to an error in the factor itself, unlike the standard error and mean absolute error which represent the error in the calculated acidifying impact. Within LCA this approach has been used before in the EDIP2003 methodology to determine an error in the site-generic AF using site-dependent AFs (Hauschild & Potting, 2005).

Table 4.3 gives one example of the results. Since in the calculation of the weighted standard deviation the weighted average is used, this value is also listed. Actually the weighted standard deviation relates to using the weighted average AF instead of sector specific AFs. In the given example the weighted average AF(-50%) and AF1 about equal the national AF (compare last two rows of the first two columns in the table). Thus in this case the weighted standard deviation can serve as measure for the error when using national instead of sector specific AFs. However, this is not the case for all acidification factors (in the example not for AF2 and AF3) and not for all examples. A second example in Table 4.4 considers NO\textsubscript{x} from the Netherlands. As explained in section 4.1 this case results in large differences between the national and sector specific AFs. The weighted average AFs are not similar to the national AFs. Therefore, in this case, the weighted standard deviation is not a good measure for the error.

Table 4.3: values of sector specific acidification factors for SO\textsubscript{2} from NL. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>sector</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>15.17</td>
<td>13.45</td>
<td>12.25</td>
<td>21.79</td>
</tr>
<tr>
<td>sector 2</td>
<td>10.29</td>
<td>26.17</td>
<td>9.34</td>
<td>143.76</td>
</tr>
<tr>
<td>sector 3</td>
<td>10.86</td>
<td>15.98</td>
<td>10.31</td>
<td>55.51</td>
</tr>
<tr>
<td>sector 4</td>
<td>13.74</td>
<td>15.43</td>
<td>13.86</td>
<td>26.40</td>
</tr>
<tr>
<td>sector 7</td>
<td>9.40</td>
<td>22.29</td>
<td>9.42</td>
<td>111.96</td>
</tr>
<tr>
<td>sector 8</td>
<td>11.87</td>
<td>13.76</td>
<td>11.69</td>
<td>28.27</td>
</tr>
<tr>
<td>sector 9</td>
<td>17.79</td>
<td>22.74</td>
<td>23.47</td>
<td>18.70</td>
</tr>
<tr>
<td>weighted standard deviation</td>
<td>2.34</td>
<td>3.41</td>
<td>1.87</td>
<td>33.75</td>
</tr>
<tr>
<td>weighted average</td>
<td>13.36</td>
<td>14.95</td>
<td>12.30</td>
<td>33.45</td>
</tr>
<tr>
<td>national</td>
<td>13.91</td>
<td>15.08</td>
<td>14.83</td>
<td>16.68</td>
</tr>
</tbody>
</table>

Figure 4.6: standard error relative to the national AF. The error relates to the deviation of the sector specific results from the complete model relative to the acidification calculated using the national AF (see text).
Table 4.4: values of sector specific acidification factors for NOx from NL. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>sector</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>7.77</td>
<td>8.17</td>
<td>7.84</td>
<td>10.29</td>
</tr>
<tr>
<td>sector 2</td>
<td>6.13</td>
<td>7.31</td>
<td>5.96</td>
<td>16.63</td>
</tr>
<tr>
<td>sector 3</td>
<td>6.14</td>
<td>7.22</td>
<td>6.15</td>
<td>14.67</td>
</tr>
<tr>
<td>sector 4</td>
<td>6.91</td>
<td>7.09</td>
<td>7.06</td>
<td>7.32</td>
</tr>
<tr>
<td>sector 7</td>
<td>9.48</td>
<td>10.14</td>
<td>9.92</td>
<td>11.52</td>
</tr>
<tr>
<td>sector 8</td>
<td>8.58</td>
<td>8.34</td>
<td>8.15</td>
<td>9.54</td>
</tr>
<tr>
<td>sector 9</td>
<td>14.85</td>
<td>14.03</td>
<td>15.11</td>
<td>6.31</td>
</tr>
</tbody>
</table>

weighted standard deviation: 1.66 1.61 1.95 2.78
weighted average: 8.49 8.95 8.59 11.35
national: 20.08 12.90 12.89 12.70

Figure 4.7 shows for each of the six examples and for all four AFs the relative error when using the weighted mean AF instead of the sector specific mean. This relative error is calculated as the quotient of the weighted standard deviation (SD) and the weighted mean. These values are taken from Table 4.3, Table 4.4, and the tables in Appendix 5.3. From Figure 4.7 one can conclude that the error from using weighted mean AFs instead of sectoral AFs is especially large for NH3 emission changes. This is probably related to the very small NH3 emission for all sectors except agriculture and the corresponding step-wise ∆A(∆E) graph. For most examples (all but NH3 from NL) AF3 shows the largest error.

![Figure 4.7: relative measure of error in AFs when considering national AFs instead of sector specific AFs.](image)

Comparing the relative error (Figure 4.7) of AF(-50%) and AF1 does not reveal a clear preference for one or the other calculation method of AFs. For AFs relates to SO2 and NH3 emissions AF(-50%) gives the lowest error while for AFs related to NOx emissions AF1 performs best (considering these two sample countries).

### 4.4 Summary and conclusions

Sector specific AFs can deviate considerably from national AFs, especially for sectors with relatively low emissions. It is however questionable to what extent the current model is suitable for the calculation of AFs based on very low emission values. In such cases the ∆A(∆E) graph consist of only a few “steps” and a linear approximation is not obvious.

---

6 Although we would prefer a measure relative to the previously determined national AF, no method to do so has been found.
To quantify the error from using national AFs instead of sector specific AFs, this chapter introduced two methods. The first method results in an error in the calculated acidifying impact when using sector specific emissions in combination with national AFs. This error combines the error from using the “wrong” AF with the linearization error. The magnitude of this error strongly relates to the emission values, making comparisons more difficult. A second method uses the weighted standard deviation of sectoral AFs. This error seems largest for NH$_3$ emissions. For other substances this error is less than 25% for most AFs. However, the weighted standard deviation is a measure relative to the weighted average of sector specific AFs and not to the national AFs, as would be preferable.

The analysis of sector specific results showed a slight preference for regression techniques to calculate AFs. AF3 seems most sensitive for sectoral differences. The size of “jumps” in the $\Delta A(\Delta E)$ graph and the corresponding emission change have a great impact on the value of AFs and the corresponding error.

If the emissions are known per sector, a sectoral approach to calculate the acidifying impact will increase the accuracy of the result. However, some sector specific AFs might be less reliable because they are based on $\Delta A(\Delta E)$ graphs that show the limitations of the model.
5 Results from grid cell specific emission changes

After having analysed the way AFs depend on the sector the emissions take place in, this chapter focuses on grid cell specific AFs. When the location of the emission change is known in more detail than on country or sector level, it is possible to use grid cell specific AFs. Such AFs will result in even more accurate acidification calculations.

This chapter considers the same examples as the previous chapter: SO$_2$, NO$_x$, and NH$_3$ emissions from France and the Netherlands with an emission and deposition resolution of 50x50km$^2$. Figure 5.1 and Figure 5.2 show the location of the grid cells that are used for the analyses in this chapter.

Figure 5.1: selected grid cells in the Netherlands

Figure 5.2: selected grid cells in France

Figure 5.3 visualizes that, in general, emissions per grid cell have lower values in France than in the Netherlands. The small emission amounts per grid cell, and corresponding very small magnitude of grid cell specific emission change ($\Delta E$), will probably cause hardly any change in area of protected ecosystem in Europe. Unlike in the previous chapter, the grid cell specific emissions do not add up to the national emissions since we only consider a few grid cells, not all grid cells in the countries.
Figure 5.3: emissions from each of the considered grid cells.

5.1 $\Delta A(\Delta E)$ graphs

Figure 5.4 and Figure 5.5 depict two examples of grid cell specific $\Delta A(\Delta E)$ graphs (Appendix 6 shows the other 4 examples). In the first figure the effect of very small emissions changes is visible. Changes in the very small amount of SO$_2$ emissions from grid cell 4 in France even cause no change at all in area of unprotected ecosystem in Europe. Reduction up to 50% of SO$_2$ emissions in grid cell 1 and 3 in France only once cause a (small) change in area of unprotected ecosystem in Europe. The emission changes still cause (very small) changes in the deposition map, but none of the European ecosystems changes its state. The used approach is not capable of handling such small changes. The same effect was already visible in the previous chapter on sector specificity, but for grid cell specificity the effect is even more dominant. The grid cell specific NO$_x$ emissions from the Netherlands are larger (see Figure 5.3) therefore the effect is not observed in Figure 5.5.

Figure 5.4: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from France. The national total (top left graph) is compared to grid cell specific results.
A possible solution to this problem could be a further refinement of the model. When an even finer deposition resolution is chosen, the deposition peak can be detected more accurately. Figure 5.6 gives a very simplified graph of the deposition versus the distance from a point source. Dots (●) represent the coarse receptor grid. The peak of the deposition is not “caught” by this grid. However, when a much finer receptor grid is applied (crosses in the figure) the peak is caught. The deposition at those receptor points can be higher than that of the receptor points related to the coarse resolution. The middle graph of the figure depicts in one dimension the grid cell deposition when the coarse receptor resolution is used while the right graph depicts the deposition on a finer receptor grid. Some locations receive a higher deposition at the finer receptor resolution, for example at a distance 4 from the emission source (compare value at distance of 4 for the middle and right graph). An ecosystem within the corresponding small grid cell might just change its state now while it didn’t when in the coarse grid cell. The described refinement of the model enables the handling of smaller emission changes.

Figure 5.5: ΔA(ΔE) graphs for NO$_x$ emissions from the Netherlands. The national total (top left graph) is compared to grid cell specific results.

Figure 5.6: simplified graphs of deposition versus distance from the emission source. The left graph is the exact result while the middle and right graph represent the deposition in discrete receptor grid cells.
However, to refine the model in such a way not only the deposition resolution should be refined, the critical load database should be refined as well. The grid cells of the database should be of the same size as the deposition grid. This also means that the maximum size of ecosystems reduces to this grid cell dimensions. Thus, the information inside the database (ecosystem area and critical load function) should be refined as well. The emission and deposition resolution of Eutrend can be refined relatively easy, but refining the critical load database is much more difficult. For a large part of Europe ecosystem information is not available on a finer scale than 50x50km$^2$.

5.2 Value of AFs

Although the $\Delta A(\Delta E)$ graphs in Figure 5.5, Figure 5.4, and Appendix 6 indicate that the model used does not produce suitable results for the small emission changes related to grid cell specific emissions, acidification factors have been calculated anyway (where possible). Figure 5.7 shows the resulting values for the AFs while Figure 5.8 shows the deviation of grid cell specific AFs from national AFs.

Figure 5.7: values of national and grid cell specific acidification factors.

Like in the previous analysis, it seems that the magnitude of grid cell emissions strongly influence the AF. Compared to Figure 4.5 the results in Figure 5.8 deviate even more from the national AFs. AFs seem to depend more heavily on the grid cell than on the sector. This effect is (among other things) related to the larger emissions per sector compared to the emissions per grid cell.

Figure 5.8: deviation of the grid cell specific AFs of Figure 5.7 from national AFs.
The selection of the preferred method to calculate AFs also suffers from the model limitations, especially the examples from France. It is therefore impossible to show a preference based on grid cell specific sensitivities. The same holds for comparing overall AFs with emission range specific AFs.

5.3 Error

The error from using grid cell specific AFs instead of the complete model applied to grid cell specific emissions suffers from the same problem as the errors related to sectoral AFs. Since only small ranges of $\Delta A$ correspond to the changing grid cell emission (see Figure 5.5, Figure 5.4, and Appendix 6), the linearization error is also very small. Both $\hat{\sigma}$ and $U$ are inadequate for comparing national and grid cell specific results. For completeness Appendix 6.2 depicts the different grid cell specific errors without a comparison to the national errors. However, the results are strongly influenced by the model’s limits and are therefore not very reliable and thus not further discussed here.

5.4 Summary and conclusions

The main conclusion from this chapter is that the limits of the model are reached when grid cell specific calculation are performed. It is therefore not possible to draw sensible conclusions considering grid cell specific AFs versus national AFs.
6 Results from other sensitivity analyses

The previous three chapters dealt with the analyses considering spatial parameters. They indicate the influence of the spatial resolution of emission and deposition, the spatial distribution of emission changes and the actual spatial location of emission changes on AFs. The current chapter discusses two other sensitivities of AFs. They are compared to the sensitivity for spatial parameters to determine a ranking of sensitivities.

Like for the analysis of sector and grid cell specific emission changes, this chapter uses SO\(_2\), NO\(_x\), and NH\(_3\) emissions from the Netherlands and France as the six examples.

6.1 Numerical resolution

The numerical resolution is a model property that will influence the calculated AFs. At two different locations in the modelling process the numerical resolution is decreased. The first analysis considers a decreased numerical resolution of the European deposition maps calculated by Eutrend (using national emission files). The second analysis intervenes later in the modelling process: the resolution of the deposition maps that serve as input for the critical load program is reduced.

6.1.1 Eutrend numerical resolution

Eutrend version 1.19 was compiled especially for the current research. It produces deposition maps in which the deposition is given with a higher numerical resolution (6 digits). This high numerical resolution does not have any physical meaning but was included to reduce the influence of numerical resolution in the current research. The original Eutrend version 1.18 has a much smaller numerical resolution (3 digits). All other programs in the modelling procedure use the original (high) numerical resolution.

The influence of the numerical resolution of Eutrend is illustrated with the $\Delta A(\Delta E)$ graphs for SO\(_2\) emissions from France and the Netherlands (Figure 6.1 and Figure 6.2). The graph for France is almost independent of the numerical resolution, as is the case for all other examples (not shown here). No explanation can be given for the exceptional behaviour of SO\(_2\) from the Netherlands.

Figure 6.3 depicts the sensitivity of AFs for the numerical resolution of the output of Eutrend. In general this sensitivity is much lower than the sensitivity of AFs on spatial parameters. Compared to Figure 3.10, Figure 4.5, and Figure 5.8 the dependence of AFs on Eutrend’s output numerical resolution (Figure 6.3) is only minor. For five of our six examples AFs vary less than 10% when the numerical output resolution of Eutrend is changed.
Figure 6.1: $\Delta A(\Delta E)$ graphs of SO$_2$ emissions from France when using Eutrend 1.19 (left) and Eutrend 1.18 (right).

Figure 6.2: $\Delta A(\Delta E)$ graphs of SO$_2$ emissions from the Netherlands when using Eutrend 1.19 (left) and Eutrend 1.18 (right).

Figure 6.3: relative change in AFs when using Eutrend 1.18 (low numerical resolution) instead of Eutrend 1.19 (high numerical resolution).
6.1.2 Deposition map resolution
In a second experiment considering numerical resolution we used Eutrend 1.19 to calculated deposition maps. However, before applying the critical load program we reduce the numerical resolution of the deposition map to three digits.

Figure 6.4 shows the sensitivity of AF for this way of limiting the numerical resolution. For a few AFs ($AF_1$ and $AF_2$ related to NO$_x$ from NL, $AF(-50\%)$ related to NO$_x$ from FR, and $AF_3$ related to NH$_3$ from NL) the deviation is more than 10%. All other AFs show lower sensitivities. Also this kind of numerical resolution limitation causes a lower sensitivity than the spatial parameters.

![Figure 6.4: relative change in AFs when changing the numerical resolution of the deposition maps before applying the critical load program.](image)

6.2 Critical load database
A second analysis focuses on the critical load database. First the influence of the (spatial) resolution is considered and second the sensitivity of AFs for the database’s base year is studied.

6.2.1 Resolution
The critical load program allows the usage of either the 50x50km$^2$ or the 150x150km$^2$ critical load database in combination with 150x150km$^2$ deposition maps. We used this property to study the influence of the critical load data base resolution. Figure 6.5 depicts changes in acidification factors calculated with 50x50km$^2$ and 150x150km$^2$ critical load database. The resolution of the critical load database hardly influences the value of the AFs.
**Difference between 50x50km² and 150x150km² critical load database**

The 50x50km² critical load database has a 2.1% larger total area of ecosystems in Europe than the 150x150km² database. Also the number of ecosystems within the database is larger, the difference is 0.25%. These differences can be explained by the way the database is filled. If a grid cell is part of a country which has submitted data, no EU background data will be used in that grid cell. EU background data provides critical load information for countries that have not submitted data. Figure 6.6 shows in what way this effect influences the total area of ecosystems per grid cell. Within the concerned 150x150km² grid cell the part covered by countries that have submitted data (grey) is also covered by 50x50km² grid cells. The other area of this 150x150km² grid cell is not accounted for in the 150x150km² database. However, in the 50x50km² database this area can have been filled with EU background data. Because of this effect the total area of ecosystems within a 150x150km² grid cell will always be equal to or less than the sum of total area of ecosystems within the nine corresponding 50x50km² grid cells. The example of the figure shows that the 150x150 grid cell contains 5+3+18+1+4=31 ecosystems while the sum of the ecosystems in the corresponding nine 50x50 grid cells might be larger than 31 because four grid cells could have been filled with background data.

![Figure 6.6: illustration of definition of number of ecosystems per grid cell. (See text)](image)

**6.2.2 Base year**

For all previous results we applied the 2004 critical load database. In this experiment we switched to the 2003 database. Differences between these two databases are defined in the CCE progress report (Hettelingh et al., 2004b).

---

Figure 6.7 and Figure 6.8 show two examples of difference in the $\Delta A(\Delta E)$ graph caused by the base year of the critical load database. In contrast to differences caused by numerical deposition resolution (e.g. Figure 6.1) differences caused by different base years of the critical load database are clearly visible (right and left graphs of Figure 6.7 and Figure 6.8 are distinguishable).

Figure 6.7: $\Delta A(\Delta E)$ graphs of NO$_x$ emissions from the Netherlands when using the 2004 (left) or 2003 (right) critical load database.

Figure 6.8: $\Delta A(\Delta E)$ graphs of NO$_x$ emissions from France when using the 2004 (left) or 2003 (right) critical load database.

Differences in $\Delta A(\Delta E)$ graphs cause differences in AFs, as depicted in Figure 6.9. The large difference in AF(-50%) for NO$_x$ emissions from the Netherlands clearly follows from Figure 6.7. The large “jump” at high emissions reductions causes $\Delta A(-50\%)$ to be much larger in the left graph of Figure 6.7 than in the right graph. The same effect was described before in section 4.2.

From Figure 6.9 follows a slight preference of using regression AFs instead of AFs based on the SV method. For all six examples $AF(-50\%)$ is more sensitive for the base year of the database than $AF1$. 
6.3 Summary and conclusions

Both analyses of the influence of numerical deposition resolution on AFs show that this parameter has less influence on AFs than the spatial parameters. Comparing the two analysis shows that the stage within the modelling process where the numerical resolution change takes place is important: Figure 6.3 and Figure 6.4 show different patterns.

The sensitivity of AF to the base year of the critical load database (Figure 6.9) is larger than to the resolution of the database (Figure 6.5). The sensitivity to the numerical resolution of the depositions (Figure 6.3 and Figure 6.4) is situated in between. However, all these influences are much smaller than those of the spatial resolution (Figure 3.10). Therefore, when extending the application area of AFs, spatial resolution seems the most important to consider.
7 Discussion

In the previous chapters the results were presented structured according to the columns of Table 2.5. This discussion chapter uses the other dimension: sections are organised according to the rows of Table 2.5. Each kind of analysis: \( \Delta A(\Delta E) \) graphs, variability of AF, values of AF, and linearization errors, is discussed in a separate section. The last section of this chapter deals with future research challenges.

7.1 \( \Delta A(\Delta E) \) graphs

Unprotected/protected ecosystems

The basis of the analysis in this report is a model that determines which European ecosystems become protected for acidification when a certain country reduces its emissions. The model’s output is a \( \Delta A(\Delta E) \) graph in which changes in protected European ecosystem area (\( \Delta A \)) are given as a function of national (sectoral, grid specific) emission changes (\( \Delta E \)). Emissions change with steps of 1%. The corresponding steps in \( \Delta A \) are of varying size.

The size of \( \Delta A \)-steps is determined by the number of ecosystems changing their state (protected/unprotected) and the area of these ecosystems. The smallest step size equals zero. This occurs when the change in deposition does not cause a change of the state of any of the European ecosystems. This lack of effect in an ecosystem can have three origins (assuming an emission reduction):

1. Small deposition reductions can be insufficient to flip an ecosystem from protected to unprotected.
2. Ecosystems can be highly exceeding their critical load and requiring a very large deposition reduction to change their state.
3. The ecosystem was already protected.

When the \( \Delta A \)-step is not zero, the minimum situation is that one European ecosystem changes its state. In that situation \( \Delta A \) equals the area of this single ecosystem. The smallest \( \Delta A \)-step (besides zero) is therefore given by the smallest ecosystem area of all ecosystems included in the database.

In practical situations the \( \Delta A \)-step seldom relates to one single ecosystem changing its state. The number of ecosystems in a grid cell that change their state depends on the deposition change in the grid cell. All ecosystems within the grid cell experience the same deposition change. Refining the size of the grid cells, and at the same time increasing the deposition resolution to the same level, initiates a spread in ecosystems changing their state. The deposition resolution should match the spatial resolution of the critical load database. The use of ecosystem specific deposition within the EMEP grid cell, as realized in the new EMEP model (Tarrasón et al., 2004), is another way of refining the deposition resolution in combination with the resolution of the critical load database.

Refining the model in the way described in the previous paragraph will not only influence the size of the \( \Delta A \)-step but also the \( \Delta E \) at which an ecosystem changes its state. National emission changes cause nearby depositions to change more than remote depositions. Ecosystems in the neighbourhood of the emission source experience a relatively large deposition change. Therefore, even ecosystems that are rather heavily exceeding their critical load can become protected when nearby emission reductions take place. Ecosystems at a larger distance from the emission source will experience a smaller change.
in deposition. Therefore, only remote ecosystems that are slightly exceeding their critical load function can become protected from these emission reductions. This theory led to the suggestion to only include local deposions. However, most examples in the current research show some remote ecosystems changing their state. These results indicate that remote ecosystems cannot be left out of the analysis. It is important to include a large part of Europe as deposition area, not just areas close to the emission sources.

Unprotected remote ecosystems (far away from the emission source) only become protected when either the emission (and thus deposition) reduction is large or when the ecosystem is only slightly exceeding its critical load function. Assuming an equal distribution of exceedances, ecosystems in the neighbourhood of the changing emission source are more likely to change their state. This situation would result in a circle-symmetric pattern (around the emission source) of ecosystems changing their state. The number of ecosystems changing their state would decrease with increasing distance from the emission source. However, such a pattern was not observed. Ecosystems in the northern part of Europe are more often unprotected than those in the southern part (Amann et al., 2004). The prevailing wind direction in middle Europe is from south-west to north-east (Maas et al., 2004), blowing emissions towards the north-eastern part of Europe. These two facts explain why we observed the majority of ecosystem changes to take place north-east of the emission sources.

“Jumps” in $\Delta A(\Delta E)$ graphs

Each emission change of 1% causes a $\Delta A$-step. $\Delta A$-steps which are considerably larger than average are called “jumps”. These “jumps” make the $\Delta A(\Delta E)$ graphs look discontinuous (in fact the graphs are always discontinuous as explained in the previous section). “Jumps” seem to be the major cause of variations in acidification factors. We observed that the magnitude of “jumps” is often dominated by one large ecosystem changing its state. Reducing the magnitude of “jumps” therefore would relate to reducing the maximum size of an ecosystem in the database. This measure would reduce the discontinuity of the $\Delta A(\Delta E)$ graph and therewith reduce the variation in AF.

In our examples the dominating ecosystem of the “jumps” is often found far away from the emission source. However, this probably relates to the choice of sample countries. Within the critical load database the considered emitting countries themselves have a large number of relatively small ecosystems, so local deposition changes will cause small $\Delta A$.

Besides the magnitude of the “jumps” also the $\Delta E$ where they occur are of importance. When they occur at small values of $\Delta E$ their influence on AFs is larger. However, no explanation of the $\Delta E$ where “jumps” occur can be given since “jumps” are just ecosystems changing their state and these ecosystems happen to be relatively large.

Importance for air pollution policies

Since the introduction of effect-based approaches within the Convention on Long-range Transboundary Air Pollution, the critical load approach has been applied for modelling exercises to support negotiations. The Convention does not (yet) use AFs but results related to $\Delta A(\Delta E)$ graphs are of importance. The non-systematic dependence of these graphs on spatial emission/deposition resolution and other model parameters show that modelling results are sensitive for these parameters. Refining the spatial resolution of the model leads to unpredictable variations in the results, as has happened at the introduction
of the new EMEP model. Also changes in the critical load database have previously caused varying results (Tarrasón et al., 2004). The current research showed similar effects. The sensitivity of the model for spatial emission and deposition resolutions raises the question of an optimal resolution. It is unknown which emission and deposition resolution gives most accurate results. Model results of different resolutions might be averaged to reduce uncertainty in the model results with respect to resolution. This approach is similar to the proposed use of multiple models in an ensemble to reduce model uncertainty in the results (Cocks et al., 1998).

7.2 Variability of AFs

Eutrend calculates deposition maps for different source/receptor resolutions (for all experiments we used the 50x50km² resolution of the critical load database). Unfortunately the different resolutions not only result in different distributions of deposition, but also in different aggregated total European depositions. Calculations with a finer source/receptor resolution did not reduce the variability of AFs calculated using small emission changes. We consider it very unlikely that the effect of different aggregated total European depositions counteracts the effect of a finer spatial resolution of emissions and deposition. Therefore, we conclude that the emission/deposition resolution does not cause the variability, as supposed by Hettelingh et al. (2005).

Experiments with a deposition pattern created by averaging 50x50km² resolution maps into 150x150km² maps, thereby conserving the aggregated total deposition, do not suffer from differences in aggregated total European deposition. Also these experiments show that the deposition resolution does not cause the observed variability of AFs calculated using small emission changes. These experiments also indicate that Eutrend is suitable for our study.

Cause of variable behaviour of AF

The experimental results showed that a finer resolution, either at the emission or at the deposition site, does not decrease the variability of the acidification factors (AFs) calculated using small emission changes. This paragraph gives an alternative explanation for the observed variability.

$AF(x)$ equals the slope of the line through points $(x, \Delta A(x))$ and $(0,0)$ of the $\Delta A(\Delta E)$ graph. For points $(x, \Delta A(x))$ close to the origin, the actual position very much influences the slope of the line. A small variation in the actual position results in a large variation of the slope. The hypothetical example in Figure 7.1 depicts this effect. The example considers two sample points (♦ in Figure 7.1), one with a large emission reduction ($x = -40\%$) and one with a small emission reduction ($x = -10\%$). Ideally the two sample points lay on the same line through the origin (dashed line in Figure 7.1) and therefore represent equal AFs.

Uncertainty in the $\Delta A(x)$ coordinate of the sample points introduces variations in $AF(x)$. Equal $\Delta A$ variations of the sample points introduce larger variations in $AF(x)$ for small $x$ (light grey lines) than for large $x$ (dark grey lines). The uncertainty in $\Delta A$ is related to the area of one ecosystem changing its state and it cause by the binary nature of the followed approach in which an ecosystem is either protected or unprotected. Therefore, the uncertainty in $\Delta A$ is of equal magnitude for all $\Delta E$ (equal vertical error bars in Figure 7.1). From the figure one sees that the uncertainty in $\Delta A$ causes a larger variation in slopes when
considering small \( x \) (light grey lines) than when considering large \( x \) (dark grey lines). This explains why \( AF(x) \) shows more variation for small \( x \) than for large \( x \).

![Graph](image)

Figure 7.1: (hypothetical example of) influence on slope of uncertain of sample point \( (x, \Delta A(x)) \) considering small (-10\%) and large (-40\%) values of \( x \), for equal absolute uncertainty in the sample points (●).

The same can be explained in a more mathematical way. \( AF \) follows from:

\[
AF(x) = \frac{\Delta A(x) \pm \text{uncertainty in } \Delta A(x)}{x}
\]

Equation 7.1

Or when separating the theoretical value and the uncertainty:

\[
AF(x) = \frac{\Delta A(x)}{x} \pm \frac{\text{uncertainty in } \Delta A(x)}{x}
\]

Equation 7.2

This leads to an uncertainty in \( AF(x) \) which is proportional to \( 1/x \):

\[
\text{uncertainty in } AF(x) = \frac{\text{uncertainty in } \Delta A(x)}{x} = \text{constant} \frac{1}{x}
\]

Equation 7.3

Since the uncertainty in \( AF(x) \) behaves as \( 1/x \), it is clear that is value is larger for small \( x \) than for large \( x \).

The uncertainty in \( \Delta A \), and therewith the variability of \( AF(x) \), could be reduced if the critical load database would be refined to contain smaller ecosystems with varying critical load functions. However, the binary nature of the approach where ecosystems are either protected or unprotected will always cause discrete steps and therefore variability in the AFs calculated using small emission changes (Heijungs & Huijbregts, 1999). Using a different approach that results in a continuous function of acidification versus \( \Delta E \), for example the accumulated exceedance (Seppälä et al., 2005), produces less varying acidification factors. However, also the indicator for acidification changes initiating a new discussion.

**Variability of sectoral AFs**

Relative large “jumps” in \( \Delta A(\Delta E) \) in the considered emission range (-20\% ≤ \( \Delta E \) ≤ +20\%) cause large variation in acidification factors. Such relative large “jumps” occur more easily when small emissions (and small emission changes in absolute units) are involved.
Therefore most sectoral acidification factors show a larger coefficient of variation than national acidification sectors.

7.3 Value of AFs

Calculation method for AF
In the current research we introduced regression techniques to calculate AFs as an alternative to the common SV method. Regression methods include more model samples than the SV method, thereby averaging the influence of uncertainty of the sample points. The SV method is very sensitive for uncertainties of the single value chosen to determine AF. Thus, theoretically AFs based on regression are less sensitive for small variations in the sample points. In general, experiments in this report support this property. Therefore we consider regression techniques a better way to determine AFs than the SV method.

Using regression techniques deviates from the traditional approach within LCIA. When the basic principle of marginal emission changes is accepted, AFs are (and should be) defined as the tangent of $\Delta A(\Delta E)$ at the origin. Mathematical problems applying this approach to the method that used exceedance/non-exceedance of critical loads are known (Heijungs et al., 1999).

Emission range specific or overall AFs?
For AFs based on regression the current report distinguished AFs based on samples from three emission ranges:
- $AF_1$, calculated using regression over $-50\% \leq \Delta E \leq +20\%$
- $AF_2$, calculated using regression over $-50\% \leq \Delta E \leq -20\%$
- $AF_3$, calculated using regression over $-20\% \leq \Delta E \leq +20\%$

Since integrated environmental assessment (IEA) generally involves large emission changes and LCAs usually deals with small emission changes, considering emission range specific AFs relates to studying the possible benefits and pitfalls of using LCA and IEA specific AFs.

Theoretically using emission range specific AFs increases the accuracy of the resulting acidification at the expense of having to know the emission range. Therefore it is advisable to use emission range specific AFs when the considered emission range is known. The examples used in the current research indicate a slightly increased sensitivity of $AF_3$ for model parameters. For the smaller emissions related to $AF_3$, uncertainty in $\Delta A$ has more impact than for the larger emission reductions related to $AF_2$. Therefore, using a separate AF for small $\Delta E$ ranges is only advisable when the conditions are very similar to the conditions for which the AF was derived. For the parameters considered in this report $AF_1$ and $AF_2$ show similar sensitivities. Based on these observations, no preference revealed for the use of either an overall AF ($AF_1$) or an emission range specific AF for the higher emission range ($AF_2$).

Comparison of AFs from this study with those from Hettelingh et al. (2005)
Acidification factors calculated in the current study deviate from the ones calculated in Hettelingh et al. (2005), see Table 7.1. Four apparent differences exist:
1. Hettelingh et al. (2005) used the critical load database of 2003, we used the one of 2004
2. Hettelingh et al. (2005) took emission data from Schöpp et al. (2003), we took data from the EMEP database.
3. Hettelingh et al. (2005) applied source-receptor matrices from the EMEP Lagrangian model to determine the deposition, we applied Eutrend.

4. Hettelingh et al. (2005) used an emission resolution on country level\(^7\). We mimicked this situation but the results are not very reliable.

Ad 1. The influence of the critical load database base year has been studied separately in chapter 6. A maximum change of 40% in the AFs has been observed.

Ad 2. From Potting et al. (1998a; 1998b), Krewitt et al. (2001), and Huijbregts et al. (2000) it is clear that the magnitude of base year emission influences the acidification factors. They reported factors for 2010 which deviate from the ones for 1990/1995. The last two columns of Table 7.1 indicate that the difference in emissions used in this study compared to the ones used by Hettelingh et al. (2005), does not solely cause the difference in AFs.

Ad 3. The model used to calculate the acidification factors also determines the AF values: Potting and Krewitt used different models and their factors differ. Each model determines deposition from emissions in its own way.

Ad 4. Acidification factors depend on the used spatial resolution for emissions and depositions in Eutrend in a non-systematic way. This relates to non-systematic changes in the \(\Delta A(\Delta E)\) function.

<table>
<thead>
<tr>
<th></th>
<th>Hettelingh et al.</th>
<th>this study</th>
<th>ratio of emissions</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>emission [Gg/y]</td>
<td>emission [-50%] [Gg/y]</td>
<td>AF [m(^2)/kg]</td>
</tr>
<tr>
<td>SO(_2) from DE</td>
<td>731</td>
<td>636</td>
<td>37.22</td>
</tr>
<tr>
<td>SO(_2) from FR</td>
<td>717</td>
<td>627</td>
<td>14.89</td>
</tr>
<tr>
<td>SO(_2) from IT</td>
<td>1096</td>
<td>752</td>
<td>2.97</td>
</tr>
<tr>
<td>NO(_x) from BE</td>
<td>310</td>
<td>329</td>
<td>14.89</td>
</tr>
<tr>
<td>NO(_x) from ES</td>
<td>1446</td>
<td>1333</td>
<td>1.57</td>
</tr>
<tr>
<td>NO(_x) from GB</td>
<td>1798</td>
<td>1718</td>
<td>8.84</td>
</tr>
<tr>
<td>NH(_3) from NL</td>
<td>360</td>
<td>322</td>
<td>60.46</td>
</tr>
<tr>
<td>NH(_3) from PL</td>
<td>54</td>
<td>58</td>
<td>109.36</td>
</tr>
</tbody>
</table>

The difference between AFs calculated by Hettelingh et al. (2005) and the AFs of this study rises the question of robustness of the factors. AFs seem to depend not only on model parameters but also on the models themselves. Ensemble modelling, using several models (and model parameters) to obtain a range of results, could reduce the dependence on a single model (and a single set of model parameters). This approach has for example been used in photochemical air-quality modelling (Delle Monache & Stull, 2003).

**Sector specific AFS**

Using sectoral data to model different spatial distributions introduced some extra dependences. Not only the spatial distribution differs for different sectors but also emission height, heat content, diurnal variation, and emission strength. Studying the different aspects separately would be a useful future extension. Since the parameters of the sectoral sources represent the actual sources, the calculated sectoral acidification factors are

---

\(^7\) The country to grid SRMs in RAINS do embody more detailed national emission distributions while the area sources used in Eutrend show a homogeneous distribution of the emissions.
correct. However, the translation into spatial distribution dependence can be questioned because dependence on emission amounts seems to dominate the results.

Because some sectors only emit very small amounts of a certain substance, model limitations influenced the results. For example NO\textsubscript{x} emissions from the Netherlands in the economic sector related to waste treatment and disposal, suffers from this effect (see Figure 4.2). The effect is seen as horizontal lines in the $\Delta A(\Delta E)$ graph. Small changes of emissions from these sectors are visible in the European deposition map, indicating that the dispersion and deposition model Eutrend can handle the small emission changes. However, the small deposition changes cause no change in protected ecosystem area. This part of the complete model seems the bottleneck when considering very small emission changes.

Applying national acidification factors for sector specific emission changes, introduces additional errors in the resulting acidification. Such errors are expressed as an emission weighted standard deviation of the sectoral AFs. They indicate the error made when using weighted average AFs instead of sector specific AFs in a way similar to the error introduced when using site-generic instead of site-dependent factors (Hauschild et al., 2005). The fact that the weighted average AFs sometimes do not equal national AFs remains a point of attention.

**Grid cell specific AFs**
The used model does not allow deriving grid cell specific AFs for all considered grid cells. When the grid cell specific emission is very low, the model calculates no (hardly any) effect. Deriving sensible grid cell specific AFs requires another model.

**Other dependences of AFs**
AFs slightly depend on the numerical resolution of the complete model. Differences were observed when considering the stage in the modelling process where the numerical resolution is decreased. Although the sensitivity of AF for numerical resolution is less than for spatial resolution, the effect cannot be neglected.

Updating parameters that define the critical load function of ecosystems influences the database. The area of unprotected ecosystem therefore depends on the version of the critical load database. We observed a dependence of AFs on the database base year that is in between sensitivity for spatial resolution and numerical resolution. Thus, improvements in the critical load database are an important factor when considering AFs.

**Importance for LCA**
Using sector or grid cell specific acidification factors would require extra information in the inventory table of an LCA. For each emission the sector or grid cell it originates from should be added. This disaggregation of the inventory table is comparable to the disaggregation required for site-dependent impact assessment. It requires extra effort but it increase the accuracy of the results.

Within LCA, site-dependent impact assessment still causes discussion. Introducing sectoral and/or grid cell specific factors will increase the resistance within the LCA community. They make LCAs become more and more complex. However, studying sector or grid cell specific emission changes can help to quantify the error introduced by not going into such detail. This information helps to define the accuracy of LCA results.
Importance for air pollution policies
The large dependence of the value of AFs on the emission/deposition resolution and other model parameters restricts the use of these AFs. Perhaps the use of ensemble modelling can help reducing this uncertainty within AFs. When considering emission reductions from just one economic sector, national AFs should be replaced by sector specific AFs.

7.4 Linearization errors
Errors calculated in this report relate to only one source of error: linearizing the $\Delta A(\Delta E)$ relation that followed from the complete model. The calculation of acidification factors contains much more uncertainties like uncertainties in the complete model itself and in the parameters it uses. Uncertainty of the model itself relates to assumptions and simplifications made to model the real world. Examples of parameters which contain uncertainty include emission heights, deposition velocities, or critical load data. A Finish study (Syri et al., 2000) analysed the uncertainty in critical load exceedance in that country. They distinguished uncertainty from emissions, transport, and critical loads, the last one turned out to be the most important.

Error related choice of AF
The analysis of linearization error in this report only considers error ranges equal to the regression range. Including errors related to using AFs on non-matching emission ranges would be a useful extension.

Considering the standard error, $\hat{\sigma}$, $AF1$ shows smaller errors than $AF(-50\%)$, expressing a preference for regression based AFs. The mean absolute error, $U$, does not always show the same behaviour as $\hat{\sigma}$.

For most examples $AF2$ relates to smaller errors than $AF1$. The error for $AF3$ can be even smaller, but this error is more sensitive to model parameters (as is the value of this AF). Based on these results the use of emission range specific AFs can be preferred to overall AFs.

Importance for LCA
Within LCA a lot of other uncertainties contribute to the environmental impact of a product or service. The error in the acidification factor is certainly not the most important one. In a case study Huijbregts et al. (2003) show that parameter uncertainty is the most important and that scenario and model uncertainty is less important. “In the impact assessment it is assumed that ecological processes respond in a linear manner to environmental interventions” (Huijbregts, 1998). This effect is part of model uncertainty within LCA. Thus, using AFs instead of the complete model relates to model uncertainty.

When an uncertainty analysis is part of an LCA, the error as calculated in this report might not be the most suitable choice. In that case it could be more convenient to express the error as an uncertainty range of the factor itself, with unit m$^2$/kg. This way LCA software like CMLCA deals with characterization factor uncertainty. Within CMLCA each characterization factor can be defined as (Heijungs & Frischknecht, 2005):

- mean value
- kind of distribution (normal, uniform, triangular, or lognormal)
- uncertainty ($\sigma$, width, or $\phi$)

The distribution of the acidification factors is not studied in this report. But providing such information for site-dependent acidification factors would increase the acceptability.
7.5 Future research challenges

Finer resolutions
The current research only considers a limited number of resolutions. Applying an even finer resolution will increase the reality of the modelled emission and deposition. In theory such an approach is possible with Eutrend, but practical limitations restrained us from including this approach in the study presented here:

- A finer resolution of emission sources would be required, including perhaps point sources. Such data is not available for all of Europe, but it is available, for example, for the Netherlands. However, using more detailed sources for only a part of Europe deviates from our research approach.
- Both a finer source resolution and a finer receptor resolution increase the amount of computing time. Eutrend calculates for each source the resulting deposition in each of the receptor points. Doubling the number of emission sources doubles the computing time. The same holds for the number of receptor points. Doubling both increases the computing time with a factor four. Using the 50x50km\(^2\) emission and deposition resolution Eutrend already needs a few days to calculate deposition maps for Europe (on a standard PC bought in 2004).
- Finally, critical load information for Europe is only available on the 50x50km\(^2\) EMEP grid. Each grid cell contains a large number of ecosystems with variable area and critical load data. In reality these ecosystems are distributed within the grid cell. Ideally a map of ecosystems based on their actual location, area, and shape combined with high resolution deposition maps will produce most accurate results. Again, for the Netherlands ecosystem data is available on a finer resolution. However, considering more detailed information for only a part of Europe deviates from our current research approach.

Future studies can include experiments applying locally a finer resolution and embedding the result in a courser grid. The current study indicates that effects far away from the country where the emissions take place influence the AFs. Thus, studying only local effects is not sufficient.

Some studies on finer resolutions already exist. They do not consider acidification factors but they do consider deposition and critical load calculations. A paper from the UK describes how in agricultural areas a 50x50m\(^2\) grid for NH\(_3\) deposition leads to a higher area of unprotected ecosystems compared to a 5x5km\(^2\) grid (Dragosits et al., 2002). A Finish study (Kangas & Syri, 2002) compares NO\(_x\) and NH\(_3\) deposition on the EMEP 150x150km\(^2\) grid and on a 14x14km\(^2\) grid. The resulting area of ecosystems exceeding their critical load for acidification increased when refining the deposition grid. In the Netherlands, modelled NH\(_3\) concentrations from emission resolutions of 500x500m\(^2\) compare better to the measurements than the 5x5km\(^2\) emission resolution (van Pul et al., 2004). These three studies indicate that a finer resolution gives better results (for ammonia). No limit has been found below which applying a finer resolution does no longer improve the results. Also no similar studies considering SO\(_2\) were found.

The transfer of the lagrangian EMEP model to the newer Unified EMEP model included, among other things, a transition to a finer deposition grid. Of course the two models have been compared extensively. For example the influence on ecosystem damage (Tarrasón et al., 2004) and on source-receptor matrices (Bartnicki, 1999) were subject of study. Related to the changing resolution Tarrasón et al. (2004) found: “Changes in the grid resolution of the deposition estimates results in non-systematic changes in the calculation of exceedances. In some places, deposition will increase, in some other decrease, but
averages will be the same as in the coarse resolution estimates. The effect on exceedances will not be systematic, because exceedance results will depend also on the critical loads of the actual ecosystems in the grid cells.” Since the new EMEP model includes a lot of new features, the influence of changing the resolution of the deposition grid is hard to extract. A comparison of acidification factors calculated with each of the EMEP models would not only include a deposition grid refinement but also changes related to other model improvements like ecosystem specific depositions.

**Other future research**

The current research focused on resolution aspects related to emissions, Eutrend, and depositions. The influence of the critical load database is mentioned but not fully explored. Analyses of this influence can include:

- size of ecosystems
- critical load values of ecosystems
- level of exceedance
- spread of ecosystems on Europe

Sectoral emissions not only differ in spatial distribution but also in a number of source properties. Separately studying influences which all contribute to sectoral results (related to Eutrend input requirements) comprises:

- (sectoral) emission spread
- emission height
- heat contents of emission source
- diurnal emission patterns
- emission strength

The only transport model used in the current research is Eutrend. Combining deposition maps from different models in an ensemble enables study of the influence of the transport model itself and it is a way to determine an uncertainty range in the deposition map.

Hettelingh et al. (2005) observed variable behaviour of $AF(x)$ at emissions changes less than 20%. Since this article formed the bases of the current research, expressing emission changes as percentage was adopted. Similar analyses using absolute emission values can be performed (using data from the current research).

The used error definition turned out to be unfit for all comparisons including large differences in emission magnitudes. Alternative relative error definitions can improve the results. Also producing statistical properties of AFs themselves would be a sensible extension of the current research.

The indicator for acidification is still under development. Recent publications on acidification factors introduced new indicators (Seppälä et al., 2005) (Finnveden & Måns, 2005). Also CCE explores alternative indicators for acidification. These new indicators will show different sensitivities on spatial resolution which are worthwhile to study.

These suggestions for future research all add knowledge about the uncertainty of AFs. They contribute to the final goal: defining the uncertainty range and distribution of AFs, which helps to define the usefulness of these factors both within LCIA and IEA.
8 Conclusion

The observed variable behaviour of $\Delta A/\Delta E$ (Hettelingh et al., 2005), the trigger for the current research, does not depend on the emission and deposition resolution as studied in this report: 50x50km$^2$ and 150x150km$^2$. The variability of $\Delta A/\Delta E$ for small values of $\Delta E$ seems to be caused by large discontinuities (steps) in the $\Delta A(\Delta E)$ function. Ecosystems are either protected or unprotected causing steps in the $\Delta A(\Delta E)$ function with sizes equal to ecosystem areas. Large steps often relate to just one large ecosystem changing its state. Therefore, the effect can only be reduced by decreasing the size of the ecosystems in the critical load database. However, the effect is inherent to the used definition of acidification and cannot be eliminated when using this definition. This does not mean that AFs cannot be used to approximate acidifying impacts.

Sector specific acidification factors differ from national factors. Using sectoral factors increases the accuracy of the calculated acidification. The same holds for grid cell specific factors. The current study wanted to use sectoral factors to study the influence of spatial distribution. However, the large influence of emission strength disabled studying the influence of spatial distributions. Grid cell specific factors indicate the influence of the actual location (on a grid cell level instead of country level). However, the model used in the current research turned out to be unfit to determine such AFs.

For the resolutions used it seems that spatial aspects of the model that calculates deposition on the basis of emissions are of minor importance for the variations in AF. Observed variations in AFs calculated using different spatial emission and/or deposition resolutions seem to originate in the last part of the complete model: the critical load database. The critical load data introduces non-linearities in the $\Delta A(\Delta E)$ function. Defining binary states (protected/unprotected) to ecosystems cause steps in the same function. Both effects contribute to the error that occurs when using acidification factors instead of the complete model to calculate acidification.

For LCA applications, quantification of errors in AF can contribute to the increased attention of uncertainty of LCA results. However, embedding such errors in LCA uncertainty frameworks requires a measure for the uncertainty in the AF itself and not for the calculated acidification, the approach followed in the current research. A more complete study to the uncertainties of acidification factors could reveal the uncertainty range and distribution of AFs.

Using AFs within air pollution policy is still in its infancy. The use of AFs can increase the speed of the calculation of acidification. Linearization errors calculated in this report should be compared to errors in the acidification that follow from the complete model. The importance of different error sources should be quantified to be able to rank possible linearization errors in relation to other error sources. Ensemble modelling might be used to decrease and quantify uncertainties within AFs resulting from uncertainties in the used model and model parameters.
References


Bellekom S. Note on methodology applied to study resolution aspects affecting Acidification Factors, with emphasis on spatial resolution of emission and deposition. Groningen: 2005.


Hettelingh J-P. Ecosystem Impacts: state of knowledge with focus on application in RAINS. 2005 Slide: Why impact factors...


Kersebaum KC and Wenkel KO. Modelling water and nitrogen dynamics at three different spatial scales influence of different data aggregation levels on simulation results. Nutrient Cycling in Agroecosystems 1998; 50(1-3): 313-319.


O'Neill RV and Sadwski DA. Aggregation error in ecological models. Ecological Modelling 1979; 7 91-105.


UNECE. 1979-2004 Twenty-five years of international cooperation on the convention on long-range transboundary air pollution. 2004. United Nations Economic Commission for Europe. Ref Type: Pamphlet


van Jaarsveld H. Receptor points in Eutrend using the EMEP grid. 2005. Ref Type: Personal Communication


### Appendix 1  Terminology

<table>
<thead>
<tr>
<th>Term</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>acidification, expressed as area of unprotected ecosystems in Europe</td>
</tr>
<tr>
<td>AF</td>
<td>acidification factor</td>
</tr>
<tr>
<td>AF(-50%)</td>
<td>AF calculated as $\Delta A(-50%)/\Delta E(-50%)$</td>
</tr>
<tr>
<td>AF1</td>
<td>AF calculated using regression on $\Delta A(\Delta E)$ over $-50% \leq \Delta E \leq +20%$</td>
</tr>
<tr>
<td>AF2</td>
<td>AF calculated using regression on $\Delta A(\Delta E)$ over $-50% \leq \Delta E \leq -20%$</td>
</tr>
<tr>
<td>AF3</td>
<td>AF calculated using regression on $\Delta A(\Delta E)$ over $-20% \leq \Delta E \leq +20%$</td>
</tr>
<tr>
<td>CV</td>
<td>coefficient of variation</td>
</tr>
<tr>
<td>E</td>
<td>emission of one acidifying substance in a certain country</td>
</tr>
<tr>
<td>E50D50</td>
<td>emission on 50x50km$^2$ grid cells, deposition on 50x50km$^2$ grid cells</td>
</tr>
<tr>
<td>E50D150</td>
<td>emission on 50x50km$^2$ grid cells, deposition on 150x150km$^2$ grid cells</td>
</tr>
<tr>
<td>E150D50</td>
<td>emission on 150x150km$^2$ grid cells, deposition on 50x50km$^2$ grid cells</td>
</tr>
<tr>
<td>E150D150</td>
<td>emission on 150x150km$^2$ grid cells, deposition on 150x150km$^2$ grid cells</td>
</tr>
<tr>
<td>EcD50</td>
<td>emission on country level, deposition on 50x50km$^2$ grid cells</td>
</tr>
<tr>
<td>EcD150</td>
<td>emission on country level, deposition on 150x150km$^2$ grid cells</td>
</tr>
<tr>
<td>IEA</td>
<td>integrated environmental assessment</td>
</tr>
<tr>
<td>LCA</td>
<td>life cycle assessment</td>
</tr>
<tr>
<td>LCIA</td>
<td>life cycle impact assessment</td>
</tr>
<tr>
<td>SV</td>
<td>single value (method) (defined in section 2.3.1)</td>
</tr>
<tr>
<td>$u$</td>
<td>sample linearization error $= \Delta A(\Delta E) - AF \cdot \Delta E$</td>
</tr>
<tr>
<td>$U$</td>
<td>average absolute linearization error</td>
</tr>
<tr>
<td>$\hat{\sigma}$</td>
<td>standard linearization error</td>
</tr>
</tbody>
</table>
Appendix 2  Linearization errors of the examples from Hettelingh et al. (2005)

Figure A2.1: absolute sample error from using linearizations $AF \cdot \Delta E$ of $\Delta A(\Delta E)$. Four methods of calculating $AF$ are distinguished (see chapter 2.3.2 ).
Figure A2.2: standard errors for each of the four acidification factors for each of the three error ranges.

Figure A2.3: average absolute errors for each of the four acidification factors for each of the three error ranges.
Appendix 3  Other results from spatial emission/deposition resolution analysis

Appendix 3.1  ΔA(ΔE) graphs

Figure A3.1: ΔA(ΔE) graphs for SO₂ emissions from Germany, at different emission and deposition resolutions.

Figure A3.2: ΔA(ΔE) graphs for SO₂ emissions from France, at different emission and deposition resolutions.
Figure A3.3: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from Italy, at different emission and deposition resolutions.

Figure A3.4: $\Delta A/\Delta E$ graphs for NO$_x$ emissions from Spain, at different emission and deposition resolutions.

Figure A3.5: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from Great Britain, at different emission and deposition resolutions.
Figure A3.6: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from the Netherlands, at different emission and deposition resolutions.

Figure A3.7: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from Poland, at different emission and deposition resolutions.
Figure A3.8: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from Sweden, at different emission and deposition resolutions.

Appendix 3.2 AF(x)

Figure A3.9: AF(x) for SO$_2$ emissions from Germany, at different emission and deposition resolutions.
Figure A3.10: AF(x) for SO\textsubscript{2} emissions from France, at different emission and deposition resolutions.

Figure A3.11: AF(x) for SO\textsubscript{2} emissions from Italy, at different emission and deposition resolutions.

Figure A3.12: AF(x) for NO\textsubscript{x} emissions from Spain, at different emission and deposition resolutions.
Figure A3.13: AF(x) for NO\textsubscript{x} emissions from Great Britain, at different emission and deposition resolutions.

Figure A3.14: AF(x) for NH\textsubscript{3} emissions from the Netherlands, at different emission and deposition resolutions.
Figure A3.15: AF(x) for NH₃ emissions from Poland, at different emission and deposition resolutions.

Figure A3.16: AF(x) for NH₃ emissions from Sweden, at different emission and deposition resolutions.
Appendix 3.3 Dependence of error on spatial emission/deposition resolution

Figure A3.17: change of standard error related to AF(50%), AF1, AF2, and AF3 by resolution, relative to E50D50.

Figure A3.18: change of mean absolute error related to AF(50%), AF1, AF2, and AF3 by resolution, relative to E50D50.
Appendix 4 Other results from averaged deposition resolution

(emission = 50x50km$^2$)

Appendix 4.1 $\Delta A(\Delta E)$ graphs

Figure A4.1: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from Germany, at different deposition resolutions.

Figure A4.2: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from Italy, at different deposition resolutions.

Figure A4.3: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from Belgium, at different deposition resolutions.

Figure A4.4: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from Spain, at different deposition resolutions.
Figure A4.5: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from Great Britain, at different deposition resolutions.

Figure A4.6: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from Poland, at different deposition resolutions.

Figure A4.7: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from Sweden, at different deposition resolutions.
Appendix 4.2  

Dependence of error on deposition resolution

![Graph](image-url)  

Figure A4.8: standard error versus deposition resolution (emission = 50x50km²)

![Graph](image-url)  

Figure A4.9: mean absolute error versus deposition resolution (emission = 50x50km²)
Appendix 5  Other results from sector specific emission changes

Appendix 5.1  ΔA(ΔE) graphs

Figure A5.1: ΔA(ΔE) graphs for SO$_2$ emissions from the Netherlands, for national (top left) and sector specific emissions.
Figure A5.2: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from the Netherlands, for national (top left) and sector specific emissions.

Figure A5.3: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from France for national (top left) and sector specific emissions.
Figure A5.4: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from France for national (top left) and sector specific emissions.

Figure A5.5: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from France for national (top left) and sector specific emissions.
Appendix 5.2    Dependence of error on emission sector

Figure A5.6: standard error related to \(\text{AF}(-50\%)\), \(\text{AF1}\), \(\text{AF2}\), and \(\text{AF3}\) for different sectors compared to the national AFs.

Figure A5.7: mean absolute error related to \(\text{AF}(-50\%)\), \(\text{AF1}\), \(\text{AF2}\), and \(\text{AF3}\) for different sectors compared to the national AFs.

Appendix 5.3    Sector specific AFs

Table A5.1: values of sector specific acidification factors for \(\text{NH}_3\) from NL. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>sector</th>
<th>(\text{AF}(-50%))</th>
<th>(\text{AF1})</th>
<th>(\text{AF2})</th>
<th>(\text{AF3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>16.66</td>
<td>17.69</td>
<td>17.88</td>
<td>16.63</td>
</tr>
<tr>
<td>sector 2</td>
<td>13.13</td>
<td>48.95</td>
<td>14.09</td>
<td>292.58</td>
</tr>
<tr>
<td>sector 3</td>
<td>46.32</td>
<td>42.76</td>
<td>45.87</td>
<td>19.80</td>
</tr>
<tr>
<td>sector 4</td>
<td>18.09</td>
<td>29.93</td>
<td>18.05</td>
<td>112.82</td>
</tr>
<tr>
<td>sector 7</td>
<td>16.87</td>
<td>25.12</td>
<td>18.45</td>
<td>85.32</td>
</tr>
<tr>
<td>sector 8</td>
<td>22.74</td>
<td>30.01</td>
<td>30.49</td>
<td>28.52</td>
</tr>
<tr>
<td>sector 9</td>
<td>18.52</td>
<td>23.39</td>
<td>18.82</td>
<td>55.03</td>
</tr>
<tr>
<td>sector 10</td>
<td>31.43</td>
<td>34.34</td>
<td>34.95</td>
<td>30.49</td>
</tr>
</tbody>
</table>

| weighted standard deviation | 21.89 | 16.48 | 27.14 | 125.45 |
| weighted average            | 30.01 | 33.49 | 33.18 | 36.05  |
| national                    | 32.18 | 33.34 | 33.80 | 30.44  |
Table A5.2: values of sector specific acidification factors for $\text{SO}_2$ from FR. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>Sector</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>8.28</td>
<td>8.67</td>
<td>8.28</td>
<td>11.47</td>
</tr>
<tr>
<td>sector 2</td>
<td>8.81</td>
<td>11.19</td>
<td>10.40</td>
<td>16.67</td>
</tr>
<tr>
<td>sector 3</td>
<td>7.77</td>
<td>8.93</td>
<td>8.29</td>
<td>13.34</td>
</tr>
<tr>
<td>sector 4</td>
<td>8.73</td>
<td>8.94</td>
<td>8.18</td>
<td>14.23</td>
</tr>
<tr>
<td>sector 7</td>
<td>8.70</td>
<td>10.58</td>
<td>10.53</td>
<td>11.29</td>
</tr>
<tr>
<td>sector 8</td>
<td>9.95</td>
<td>11.40</td>
<td>11.32</td>
<td>12.36</td>
</tr>
<tr>
<td>sector 9</td>
<td>8.14</td>
<td>8.58</td>
<td>9.10</td>
<td>5.23</td>
</tr>
<tr>
<td>weighted standard deviation</td>
<td>0.65</td>
<td>1.20</td>
<td>1.22</td>
<td>2.27</td>
</tr>
<tr>
<td>weighted average</td>
<td>8.35</td>
<td>9.20</td>
<td>8.66</td>
<td>12.96</td>
</tr>
<tr>
<td>national</td>
<td>9.66</td>
<td>10.26</td>
<td>9.89</td>
<td>12.94</td>
</tr>
</tbody>
</table>

Table A5.3: values of sector specific acidification factors for $\text{NO}_x$ from FR. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>Sector</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>6.42</td>
<td>6.47</td>
<td>5.84</td>
<td>10.83</td>
</tr>
<tr>
<td>sector 2</td>
<td>6.00</td>
<td>6.76</td>
<td>6.27</td>
<td>10.15</td>
</tr>
<tr>
<td>sector 3</td>
<td>5.91</td>
<td>6.65</td>
<td>6.10</td>
<td>10.48</td>
</tr>
<tr>
<td>sector 4</td>
<td>8.06</td>
<td>8.67</td>
<td>9.36</td>
<td>4.25</td>
</tr>
<tr>
<td>sector 7</td>
<td>7.12</td>
<td>7.36</td>
<td>7.26</td>
<td>8.06</td>
</tr>
<tr>
<td>sector 8</td>
<td>5.12</td>
<td>5.75</td>
<td>5.34</td>
<td>8.65</td>
</tr>
<tr>
<td>sector 9</td>
<td>5.21</td>
<td>5.25</td>
<td>5.61</td>
<td>2.66</td>
</tr>
<tr>
<td>weighted standard deviation</td>
<td>1.21</td>
<td>0.99</td>
<td>1.29</td>
<td>1.86</td>
</tr>
<tr>
<td>weighted average</td>
<td>6.50</td>
<td>6.89</td>
<td>6.62</td>
<td>8.78</td>
</tr>
<tr>
<td>national</td>
<td>9.95</td>
<td>9.28</td>
<td>9.30</td>
<td>9.02</td>
</tr>
</tbody>
</table>

Table A5.4: values of sector specific acidification factors for $\text{NH}_3$ from FR. For comparison the last row gives the national acidification factors.

<table>
<thead>
<tr>
<th>Sector</th>
<th>AF(-50%)</th>
<th>AF1</th>
<th>AF2</th>
<th>AF3</th>
</tr>
</thead>
<tbody>
<tr>
<td>sector 1</td>
<td>18.80</td>
<td>14.62</td>
<td>15.59</td>
<td>8.02</td>
</tr>
<tr>
<td>sector 2</td>
<td>35.21</td>
<td>43.37</td>
<td>45.34</td>
<td>32.19</td>
</tr>
<tr>
<td>sector 3</td>
<td>13.54</td>
<td>12.02</td>
<td>12.58</td>
<td>8.39</td>
</tr>
<tr>
<td>sector 4</td>
<td>20.60</td>
<td>23.26</td>
<td>24.54</td>
<td>15.21</td>
</tr>
<tr>
<td>sector 7</td>
<td>18.19</td>
<td>21.84</td>
<td>21.98</td>
<td>21.81</td>
</tr>
<tr>
<td>sector 8</td>
<td>15.49</td>
<td>20.95</td>
<td>20.77</td>
<td>23.47</td>
</tr>
<tr>
<td>sector 9</td>
<td>16.94</td>
<td>20.33</td>
<td>20.32</td>
<td>21.11</td>
</tr>
<tr>
<td>sector 10</td>
<td>13.17</td>
<td>16.17</td>
<td>14.96</td>
<td>24.69</td>
</tr>
<tr>
<td>weighted standard deviation</td>
<td>20.39</td>
<td>23.69</td>
<td>27.77</td>
<td>15.70</td>
</tr>
<tr>
<td>weighted average</td>
<td>13.51</td>
<td>16.54</td>
<td>15.41</td>
<td>24.48</td>
</tr>
<tr>
<td>national</td>
<td>12.83</td>
<td>16.00</td>
<td>14.77</td>
<td>24.66</td>
</tr>
</tbody>
</table>
Appendix 6  Other results from grid cell specific emission changes

Appendix 6.1  $\Delta A(\Delta E)$ graphs

Figure A6.1: $\Delta A(\Delta E)$ graphs for SO$_2$ emissions from the Netherlands. The national total (top left graph) is compared to grid cell specific results.
Figure A6.2: $\Delta A(\Delta E)$ graphs for NH$_3$ emissions from the Netherlands. The national total (top left graph) is compared to grid cell specific results.

Figure A6.3: $\Delta A(\Delta E)$ graphs for NO$_x$ emissions from France. The national total (top left graph) is compared to grid cell specific results.
Figure A6.4: ΔA(ΔE) graphs for NH₃ emissions from France. The national total (top left graph) is compared to grid cell specific results.

Appendix 6.2 Dependence of error on grid cell emissions

Figure A6.5: standard error related to AF(-50%), AF1, AF2, and AF3 for different grid cells.

Figure A6.6: mean absolute error related to AF(-50%), AF1, AF2, and AF3 for different grid cells.