

Interim Report

IR-01-020

From Economic Activities to Critical Load Exceedances in Europe – An Uncertainty Analysis of Two Scenarios of the RAINS Integrated Assessment Model

Riku Suutari (suutari@iiasa.ac.at), Markus Amann, Janusz Cofala, Zbigniew Klimont and Wolfgang Schöpp

Approved by

Markus Amann (amann@iiasa.ac.at) Transboundary Air Pollution Project

May 28, 2001

Interim Reports on work of the International Institute for Applied Systems Analysis receive only limited review. Views or opinions expressed herein do not necessarily represent those of the Institute, its National Member Organizations, or other organizations supporting the work.

Contents

1	Intro	oduction1
2	The	RAINS Model1
3	Unc	vertainties4
	3.1	Earlier Uncertainty Analysis with RAINS
	3.2	A Taxonomy of Uncertainties in the RAINS Model
	3.3	A Methodology for Assessing Uncertainties7
	3.3.	1 Definition7
	3.3.	2 Terminology7
	3.3.	3 The Basic Concept
	3.4	Uncertainty of Calculated Emissions
	3.5	Uncertainty of Calculated Deposition9
	3.6	Uncertainty of Ecosystems Protection11
4	Unc	vertainties in the RAINS Calculations of Ecosystems Protection14
	4.1	Uncertainties in Emission Estimates
	4.1.	1 Assumed Uncertainties in Input Data for SO ₂ and NO _x 15
	U	Incertainties in Activity Data
	U	Incertainties in Emission Factors
	U	Incertainties in Removal Efficiencies
	4.1.	2 Assumed Uncertainties in Input Data for NH ₃
	U	Incertainties in Activity Data
	U	Incertainties in Emission Factors
	U	Incertainties in the Reduction Efficiencies of Emission Control Techniques
	4.1.	3 Resulting Uncertainties in Emission Calculations
	S	ectoral Emission Estimates
	U	Incertainties in National Total Emissions
	4.2	Uncertainties in the Atmospheric Dispersion Calculations
	4.2.	1 Assumed Uncertainties in Input Data
	4.2.	2 Resulting Uncertainties in Atmospheric Dispersion Calculations
	4.3	Uncertainties in Ecosystems Protection

	4.3.	1 Assumed Uncertainties in Input Data	37
	4.3.2 Resulting Uncertainties in Ecosystems Protection		37
5	Disc	cussion and Conclusions	42
	5.1	General Observations about Uncertainties	42
	5.2	Uncertainty of Emission Estimates	42
	5.3	Uncertainties of Deposition Estimates	43
	5.4	Uncertainties of Estimates on Ecosystems Protection	43
	5.5	Conclusions	44
6	Refe	erences	45
7	Ann	nex: Relevant Parts from Statistical Textbooks	48

Acknowledgments

The authors gratefully acknowledge the financial support received from the United Kingdom's Department of Environment, Transport and the Regions.

We thank Maximilian Posch from the Coordination Center for Effects at RIVM for providing information on the uncertainties of critical loads and for collaborating in the methodological development.

From Economic Activities to Critical Load Exceedances in Europe – An Uncertainty Analysis of Two Scenarios of the RAINS

Integrated Assessment Model

Riku Suutari, Markus Amann, Janusz Cofala, Zbigniew Klimont and Wolfgang Schöpp

1 Introduction

In recent international negotiations integrated assessment models were used to quantify reductions of a range of pollutants required to meet politically established targets of environmental quality, e.g., in terms of acidification of ecosystems (e.g., Amann and Lutz, 2000). Without doubt considerable uncertainties exist in almost all parts of the integrated assessment frameworks, e.g., in the emission inventories, the estimates of environmental sensitivities. It is also clear that these uncertainties have a direct impact on the amount by which emissions need to be reduced in order to achieve a desired environmental target. A systematic quantification of the sensitivity of the optimization results in relation to the model and data uncertainties is complicated to do and requires substantial time and resources.

This paper analyzes the uncertainties involved in estimating the areas at risk of acidification in Europe. We explore how, as a consequence of uncertainties in the input data, the uncertainty of critical loads excess in Europe changes from 1990 to 2010. Section 2 introduces the Regional Air Pollution Information and Simulation (RAINS) model that is used as the methodological framework for the analysis. Section 3 reviews the different types of uncertainties inherent in performing calculations with the RAINS model and develops a methodology to propagate uncertainties through the entire chain of model calculations. Section 4 applies this methodology to two calculations, specifying uncertainty ranges for a number of model parameters and assessing their implication on model results. Findings are discussed in Section 5, and conclusions are drawn in Section 6.

2 The RAINS Model

The Regional Air Pollution INformation and Simulation (RAINS)-model developed at the International Institute for Applied Systems Analysis (IIASA, Laxenburg, Austria) provides a consistent framework for the analysis of emission reduction strategies, focusing on acidification, eutrophication and tropospheric ozone. RAINS includes modules for emission generation (with databases on current and future economic activities, energy consumption levels, fuel characteristics, etc.), for emission control options and costs, for atmospheric dispersion of pollutants and for environmental sensitivities (i.e., databases on critical loads). In order to create a consistent and comprehensive picture of the options for simultaneously addressing the three environmental problems (acidification, eutrophication and tropospheric

ozone), the model considers emissions of sulfur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃) and volatile organic compounds (VOC). A detailed description of the RAINS model can be found in Schöpp *et al.*, 1999. A schematic diagram of the RAINS model is displayed in Figure 2.1.

The European implementation of the RAINS model incorporates databases on energy consumption for 38 regions in Europe, distinguishing 22 categories of fuel use in six economic sectors. The time horizon extends from the year 1990 up to the year 2010 (Bertok et al., 1993). Emissions of SO₂, NO_x, NH₃ and VOC for 1990 are estimated based on information collected by the CORINAIR'90 inventory of the European Environmental Agency and on national information. Options and costs for controlling emissions of the various substances are represented in the model by considering the characteristic technical and economic features of the most important emission reduction options and technologies. Atmospheric dispersion processes over Europe for sulfur and nitrogen compounds are modeled based on results of the European EMEP model developed at the Norwegian Meteorological Institute (Barret and Sandnes, 1996). For tropospheric ozone, source-receptor relationships between the precursor emissions and the regional ozone concentrations are derived from the EMEP photo-oxidants model (Simpson, 1992, 1993). The RAINS model incorporates databases on critical loads and critical levels compiled at the Coordination Center for Effects (CCE) at the National Institute for Public Health and Environmental Protection (RIVM) in the Netherlands (Posch et al., 1999).



The RAINS Model of Acidification and Tropospheric Ozone

Figure 2.1: Schematic flowchart of the RAINS model framework

The RAINS model can be operated in the 'scenario analysis' mode, i.e., following the pathways of the emissions from their sources to their environmental impacts. In this case the model provides estimates of regional costs and environmental benefits of alternative emission control strategies. Alternatively, an 'optimization mode' is available to identify cost-optimal allocations of emission reductions in order to achieve specified air quality targets. This mode of the RAINS model was used extensively during the negotiation process of the Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone of the Convention on Long-range Transboundary Air Pollution and the proposal of the European Commission on a Directive on National Emission Ceilings (Amann and Lutz, 2000).

3 Uncertainties

3.1 Earlier Uncertainty Analysis with RAINS

Like all models, the RAINS model attempts to develop a holistic understanding of a complex reality through a variety of reductionistic steps. This simplification process is burdened with many uncertainties related to methodological issues, lack of understanding and insufficient data.

Several analyses on some aspects of uncertainty have been undertaken during the development and application of the RAINS model:

Sorensen (1994a,b) conducted a sensitivity analysis for the cost calculation routine implemented in the RAINS model and explored how such uncertainties affect the outcome of an optimization analysis. In general, quantitative optimization results were found to be sensitive to variations in the capacity utilization of boilers and in the sulfur contents of fuels. While such variations might change results for individual countries, overall optimized patterns of required emission reductions, however, do not change significantly.

Altman *et al.* (1996) analyzed the influence of uncertainties in emission control costs on calculations of cost-effective European sulfur emission reductions. A specialized solution procedure was developed and a number of different cost curves were generated to model the uncertain costs.

An analysis of the robustness of RAINS-type cost curves (Duerinck, 2000) suggested that uncertainties in the cost components, although relatively high, were much less important for the overall uncertainty than uncertainties in the emissions.

The relationship between deposition targets and the calculated emission ceilings for Denmark has been investigated using Monte Carlo simulation (Bak and Tybirk, 1998). In addition, the sensitivity of the calculated emission ceilings with respect to changes in Danish national data has been analyzed. The analysis explored the sensitivity towards modifications in the energy scenario, the agricultural scenario, the ammonia emission factors and the marginal costs of SO_2 , NO_x and ammonia abatement.

Alcamo *et al.* (1987) explored to what extent interregional transport of air pollutants in Europe could be described by linear relations. It was found that the linearity between emissions and deposition strongly depends on the distance between emitter and receptor, the averaging period, the constituent (acidity, oxidants, sulfur, etc.), and the form of deposition (e.g., whether total deposition is considered or wet deposition alone).

The same authors addressed the uncertainty of atmospheric source-receptor relationships for sulfur within Europe (Alcamo and Bartnicki, 1990). Stochastic simulation was used to compute the effect on selected transfer coefficients of uncertainties related to transport wind, meteorological forcing functions, model parameters and the spatial distribution of emissions. Uncertainty estimates for 30 source-receptor combinations – based on one year's meteorological conditions – suggested a relative uncertainty of 10 percent to 30 percent in the transfer coefficients, not correlated with the distance between emission source and receptor. However, their absolute uncertainty (standard deviation) was found strongly correlated with distance and proportional to the values of the transfer coefficients themselves.

Hettelingh (1989) addressed the uncertainty of modeling regional environmental impacts caused by imperfect compatibility of models and available measurement data. He concluded that an uncertainty analysis of integrated environmental models, which integrates different processes (e.g., meteorological, soil and watershed acidification processes) with a probabilistic interpretation of model predictions, might allow different models and data to provide overlapping confidence intervals.

The uncertainty in ecosystem protection levels in Finland was found (Syri, Suutari and Posch, 2000) to be dominated by the uncertainties in critical loads for most parts of the country.

Van Sluijs (1996) compared different approaches to the management of uncertainties taken by regional integrated assessment models for climate change and regional air quality. A comprehensive treatment of uncertainties turned out to be a challenge for all models available at that time: (i) Models do not fully address all relevant aspects within the whole spectrum of types and sources of uncertainty; (ii) they failed to provide unambiguous comprehensive insight to both the modeler and the user into the quality and limitations of models and their answers and (iii) they failed to address the subjective component in the appraisal of uncertainties.

This finding did not come as a surprise to the developers of integrated assessment models, since it demonstrated that, due to the complexity of such models, an appropriate treatment of uncertainties is far from trivial.

For the particular case of the RAINS model, uncertainty was raised as a matter of concern by industry and countries when the RAINS model was used to guide negotiations under the Convention on Long-range Transboundary Air Pollution on the Gothenburg Protocol and the proposal of the Commission of the European Union on a Directive on National Emission Ceilings. The above studies provide only partial answers, since they addressed uncertainties of individual components of the overall model system.

The computational complexity of the RAINS model system made it difficult to conduct a formal uncertainty analysis with traditional approaches that would yield quantitative insight. For many of the input elements of the model insufficient quantitative information on input data uncertainties is available. Assumptions about error distributions and independence of parameters would be required; and such assumptions would themselves constitute further sources of uncertainty. Instead, the model developers decided to consider uncertainty management as an important guiding principle already during the model development phase and adopted a variety of measures in model design and scenario planning to systematically minimize the potential influence of uncertainties on policy-relevant model output (Schöpp et al., 2001). For instance, at all phases of model development and use, explicit confidence intervals (for emission control potentials, deposition ranges, ozone levels, ecosystems sensitivities, etc.) defined the range within the model was proven to work with sufficient accuracy. Potential reliance of optimized solutions on single point estimates were avoided through integral measures for environmental sensitivities. Specially designed compensation mechanisms allowed controlled violation of environmental targets for single ecosystems with potentially uncertain sensitivities. Wherever possible, preference was given to relative model outcomes (comparing two model outputs) rather than to absolute values. For ground-level ozone, less weight was given to extreme meteorological situations because their representativeness was questionable and the performance of the meteorological model for such rare situations was less certain. Sensitivity analysis attempted to identify systematic biases and showed that with large probability the emission reductions resulting from the model calculations could be considered as minimum requirements, suggesting that there is only little chance that policy measures suggested by the model needed to be revised in the future in the light of new information.

3.2 A Taxonomy of Uncertainties in the RAINS Model

To help make the assembling of uncertainties more systematic, we propose the following taxonomy, which classifies uncertainties in terms of model characteristics (after Alcamo and Bartnicki, 1987):

 Model structure – these are uncertainties resulting from the specified collection of model terms and how they are related, containing all physical assumptions of the model;

- parameters uncertainties from coefficients which are constant in time or space;
- forcing functions uncertainties from coefficients which inherently change in time and space;
- initial state uncertainties inherent in boundary and initial conditions.

Table 3.1 lists examples of these types of uncertainties, distinguishing the three major components of the RAINS model (emission calculation, atmospheric dispersion, critical loads).

Model structure	Emission calculations	Selected sectoral aggregation
		Determination of mean values
	Atmospheric dispersion	Linearity in atmospheric dispersion
		Selected spatial resolution, ignoring in-grid variability
		Country size (country-to-grid)
	Critical loads estimates	The threshold concept, e.g., the critical Ca/Al ratio
		Selected aggregation of ecosystems
		Static representation of a dynamic process
Parameters	Emission calculations	Expected values for fuel quality, removal efficiencies and application rates
	Atmospheric dispersion	Expected values of parameters for describing chemical and physical processes (conversion rates, deposition rates)
		Mean transfer coefficient in view of inter- annual meteorological variability
	Critical loads estimates	Expected values of base cation deposition and uptake, throughflow, nitrogen uptake in critical loads calculations
Forcing functions	Emission calculations	Accuracy of statistical information on economic activities
		Projections of sectoral economic activities
		Future implementation of emission controls
	Atmospheric dispersion	Spatial distribution of emissions within countries
		Accuracy of meteorological data
Initial state	Emission calculations	Uncontrolled emission factors
		State of emission controls in the base year
	Atmospheric dispersion	Natural emissions
		Hemispheric background

Table 3.1: Taxonomy of uncertainties in the RAINS model

3.3 A Methodology for Assessing Uncertainties

We propose a methodology to address the uncertainties in the various model terms (variables and parameters) and to explore how they propagate through the entire model chain (Figure 3.1) and thereby influence the uncertainty of intermediate model output, such as national emissions, deposition fields and critical load excess.



Figure 3.1: Flowchart of the uncertainty analysis for ecosystems protection. Uncertainty ranges are specified for the input parameters in the emissions, deposition and critical loads models, as well as estimating the errors from the model integration.

3.3.1 Definition

For this purpose, let's consider a deterministic model term A (either a model variable or parameter), whose uncertainty is represented by an uncertainty factor χ with the expected value of one and the standard deviation σ_{χ} , so that the uncertain model term is defined as A_{χ} . Thereby the coefficient of variation of the uncertain term A_{χ} is σ_{χ} :

$$CV_{A\chi} = \frac{\sigma_{A\chi}}{A} = \frac{A\sigma_{\chi}}{A} = \sigma_{\chi}$$
 (Equation 1)

3.3.2 Terminology

In the remainder of the paper we denote uncertain terms by Greek characters, while deterministic terms are printed in Latin. Dependent uncertain terms are marked with $\overline{\overline{X}}$; expected values of $\overline{\overline{X}}$ are marked as $\overline{\overline{X}}$. Furthermore, we denote the following statistical operators:

E[]expected valueVar[]variance of a valueCov[,]covariance of two values

3.3.3 The Basic Concept

To explore the influence of the uncertainties of individual model terms on overall model results, we have developed an analytical method to propagate the errors presented as coefficients of variation through the entire model chain. We assigned to a number of model terms uncertainty factors χ and attempted to quantify them based on available statistics and expert judgment. We determine the resulting uncertainty for the major intermediate outputs of the RAINS model, i.e., for national emissions of acidifying substances, for deposition of sulfur and nitrogen compounds and for excess deposition over critical load. Finally, using standard statistical operations we quantified the influence of these uncertainties through the entire calculation chain from emissions to ecosystems protection.

3.4 Uncertainty of Calculated Emissions

For the deterministic case the RAINS model calculates for a given economic sector j in country i the emissions $em_{x,i}$ of a certain substance x as the product of a rate of economic activity $ac_{x,ij}$ the uncontrolled emission factor $ef_{x,ij}$ (representing theoretical emissions in absence of any emission control measures), the application rates $af_{x,ij,k}$ and removal efficiencies $rem_{x,i,i,k}$ of the available emission control options k:

$$em_{x,i} = \sum_{j} act_{x,i,j} ef_{x,i,j} (1 - \sum_{k} af_{x,i,j,k} rem_{x,i,j,k})$$
(Equation 2)

Details about emission calculations are provided in Cofala and Syri (1998a,b) and Klimont *et al.*, 1998.

While this calculation formula bears uncertainties in itself (especially in cases where legally binding emission limit values are in force), all of the above mentioned model parameters are associated with uncertainties. Let's define for each of the model terms the expected value and an uncertainty factor:

act = the expected activity level

 α = uncertainty factor for the activity level

ef = expected uncontrolled emission factor per unit of activity

 \mathcal{E} = uncertainty factor for the emission factor

af = expected application rate for the abatement technology

 γ = uncertainty factor for the application rate

rem = expected removal efficiency of the abatement technology

 η = uncertainty factor for removal efficiency

With these definitions, the uncertain emissions em for pollutant x and country i are calculated as

$$\overline{\overline{em}}_{x,i} = \sum_{j} act_{x,i,j} \alpha_{x,i,j} ef_{x,i,j} \varepsilon_{x,i,j} (1 - \sum_{k} af_{x,i,j,k} \gamma_{x,i,j,k} rem_{x,i,j,k} \eta_{x,i,j,k}).$$
(Equation 3)

If we assume that no correlations exist between the uncertainty factors for the activity levels (α), the emission factors (ϵ), the application rates (γ), and the removal efficiencies (η), then the expected emissions $\overline{em}_{x,i}$ emerge from Equation 3 as

$$\overline{em}_{x,i} = \sum_{j} act_{x,i,j} ef_{x,i,j} (1 - \sum_{k} af_{x,i,j,k} rem_{x,i,j,k}), \quad (\text{Equation 4})$$

which is equal to the deterministic case (Equation 2).

However, certain variables and model parameters are common input for calculations for different pollutants (x,y), regions (i,p) and sectors (j,q) (e.g., activity rates, removal efficiencies of emission control options, etc.). Therefore, there exists a covariance between the emission estimates of different regions and of different pollutants:

$$Cov[\overline{em_{x,i}}, \overline{em_{y,p}}] = \sum_{j \in Q} (act_{x,i,j}act_{y,p,q}ef_{x,i,j}ef_{y,p,q}E[\alpha_{x,i,j}\alpha_{y,p,q}]E[\varepsilon_{x,i,j}\varepsilon_{y,p,q}]$$

$$(\sum_{j \in Q} af_{x,i,j,k}af_{y,p,q,m}rem_{x,i,j,k}rem_{y,p,q,m}E[\gamma_{x,i,j,k}\gamma_{y,p,q,m}]E[\eta_{x,i,j,k}\eta_{y,p,q,m}]$$

$$\sum_{k \in M} af_{x,i,j,k}rem_{x,i,j,k} - \sum_{m} af_{y,p,q,m}rem_{y,p,q,m}+1)) - \overline{em}_{x,i}\overline{em}_{y,p}$$
(Equation 5)

The expected value of a product is defined according to Equation 7 of the Annex.

For further analysis the variances of national emissions are also of interest. The variance of emissions of pollutant x at region i is the covariance of emission x itself

$$Var[\overline{em_{x,i}}] = Cov[\overline{em_{x,i}}, \overline{em_{x,i}}]$$
(Equation 6)

3.5 Uncertainty of Calculated Deposition

The deterministic RAINS calculations determine deposition of pollutant x at a given receptor site e by multiplying the emissions $em_{x,i}$ of all countries i with transfer (dispersion) coefficients for wet and dry deposition ($w_{x,i,e}$, $d_{x,i,e}$), respectively:

$$dep_{x,e} = \sum_{i} em_{x,i} (w_{x,i,e} + d_{x,i,e})$$
(Equation 7)

For the uncertainty analysis we associate each of the deterministic terms with an uncertainty factor and we add a term for in-grid variability (λ):

 $\xi_{x,i}$ = emission uncertainty factor for compound x and region *i*,

 $w_{x,i,e}$ = wet deposition transfer coefficient for compound x from region i to receptor site e

 $\omega_{x,i,e}$ = uncertainty factor for the wet deposition transfer coefficient

 $d_{x,i,e}$ = dry deposition transfer coefficient for compound x from region i to receptor site e

 $\delta_{x,i,e}$ = uncertainty factor for the wet deposition transfer coefficient

With these uncertainty terms we calculate deposition as

$$\overline{\overline{dep}}_{x,e} = \lambda_{x,e} + \sum_{i} \overline{em}_{x,i} \xi_{x,i} (w_{x,i,e} \omega_{x,i,e} + d_{x,i,e} \delta_{x,i,e}). \quad (\text{Equation 8})$$

A λ of zero yields the uncertainty of the average deposition in the grid cell *e*.

Because the uncertainty factors in Equation 8 have expected values of one and the in-grid variability term λ has an expected value of zero, the expected deposition \overline{dep} is

$$\overline{dep}_{x,e} = \sum_{i} \overline{em}_{x,i} (w_{x,i,e} + d_{x,i,e}).$$
(Equation 9)

Assuming the uncertainties in emissions (ξ) are uncorrelated with the uncertainties of the transfer coefficients (ω , δ), the covariance between compounds *x* and *y* in grid *e* is

$$Cov[\overline{dep}_{x,e}, \overline{dep}_{y,e}] = E[\lambda_{x,e}\lambda_{x,e}]$$

$$+ \sum_{i} \sum_{p} (\overline{em}_{x,i}, \overline{em}_{y,p}E[\xi_{x,i}\xi_{y,p}](w_{x,i,e}w_{y,p,e}E[\omega_{x,i,e}\omega_{y,p,e}])$$

$$+ \overline{dep}_{x,i,e}\overline{dep}_{y,p,e}E[\delta_{x,i,e}\delta_{y,p,e}] + w_{x,i,e}\overline{dep}_{y,p,e}E[\omega_{x,i,e}\delta_{y,p,e}]$$

$$+ w_{y,p,e}\overline{dep}_{x,i,e}E[\omega_{y,p,e}\delta_{x,i,e}]) - \overline{dep}_{x,e}\overline{dep}_{y,e}$$

(Equation 10)

where

$$\mathbb{E}[\lambda_{x,e}\lambda_{y,e}] = \sigma_{\lambda_{x,e}} \sigma_{\lambda_{y,e}} \rho_{\lambda_{x,e},\lambda_{y,e}}$$
(Equation 11)

As for emission calculations, the variance of deposition of pollutant x at the receptor site e is the covariance of deposition of the compound x itself:

$$\sigma_{x,e}^2 = Var[\overline{dep}_{x,e}] = Cov[\overline{dep}_{x,e}, \overline{dep}_{x,e}]$$
(Equation 12)

For comparing acid deposition with critical loads protection isolines, we need to combine deposition of oxidized and nitrogen compounds originating from NO_x and NH_3 emissions (Figure 3.2). The expected total nitrogen deposition is calculated as

$$\overline{dep}_{N,e} = \overline{dep}_{NOx,e} + \overline{dep}_{NHy,e}$$
(Equation 13)

The variance of total nitrogen deposition is then the sum of the variances of the oxidized and reduced nitrogen depositions and their covariance:

$$\sigma_{N,e}^{2} = \sigma_{NOx,e}^{2} + \sigma_{NHx,e}^{2} + 2\sigma_{NOx,e}\sigma_{NHx,e}\rho_{NOx,NHx,e}.$$
 (Equation 14)

The correlations ρ are derived from the covariances according to Equation 10.

The correlation between the deposition of sulfur and total nitrogen is obtained as

$$\rho_{SOx.N,e} = \rho_{SOx.NOx,e} \frac{\sigma_{NOx,e}}{\sigma_{N,e}} + \rho_{SOx.NHx,e} \frac{\sigma_{NHx,e}}{\sigma_{N,e}} .$$
(Equation 15)



Figure 3.2: Probability density of sulfur and nitrogen deposition

3.6 Uncertainty of Ecosystems Protection

The RAINS model calculates acid deposition $dep_{x,e}$ from the emissions $em_{x,i}$ and compares it with the critical loads for the ecosystems.

Deposition of nitrogen and sulfur compounds is computed as single values for each grid cell. Within a single EMEP grid cell, however, many (up to 100,000 in some cases) critical loads for various ecosystems, mostly forest soils and surface waters, have been calculated. These critical loads are sorted according to their magnitude taking into account the area of the ecosystem they represent, and the so-called cumulative distribution function is constructed. This cumulative distribution function is then compared to the single deposition values for that grid cell.

In practical terms, the RAINS model uses so-called 'protection isolines' for each grid cell describing pairs of sulfur and nitrogen deposition that protect an equal area of ecosystems. Such protection isolines are constructed by calculating protection points along rays passing through origin of the sulfur-nitrogen plane.

While the calculated deposition of sulfur and nitrogen compounds is associated with uncertainties, the protection isolines themselves also bear significant uncertainties related to the concept of establishing critical chemical thresholds, the methodology used to determine them, the underlying data, etc.

We define the uncertainties of the points of protection isolines along the rays through the origin of the sulfur/nitrogen plane (Figure 3.3), so that the probability P_n of a certain percentage of the ecosystems being protected at given deposition x is also defined along rays from the origin. The probability (Figure 3.4) is calculated for a fixed sulfur to nitrogen ratio as

$$P_n(r(x) < r(pi_n(x))) = 1 - G_n(x, r(x))$$
 (Equation 16)

where

- r(x) the distance of the point x (denoting a pair of sulfur and nitrogen deposition) from the origin (in equivalents/hectare/year), calculated as $r(x) = \sqrt{\Delta S(x)^2 + \Delta N(x)^2}$
- $r(pi_n(x))$ the distance of the isoline for protection percentage *n* from the origin for the sulfur/nitrogen ratio of point *x*
- $G_n(x, r(x))$ the value of the cumulative distribution function of the protection isoline for protection percentage *n* having the same S/N ratio as the point *x* at the distance r(x). For each S/N ratio the distribution is defined by the expected value of the distance between the isoline and the origin and the standard deviation for the same S/N ratio.



Figure 3.3: Probability density of a selected protection isoline along an arbitrary ray



Figure 3.4: Protection probability along an arbitrary ray

Evaluated for all sulfur/nitrogen ratios we obtain a protection probability surface as illustrated in Figure 3.5.



Figure 3.5: Protection probability surface for an arbitrary percentile

The uncertainties of deposition and of protection isolines can be combined, so that the probability to protect with a given deposition (D_N, D_S) n percent of ecosystems is calculated as:

$$P_n\Big|_{f_D} = \int_0^\infty \int_0^\infty P_n\big(r(D_{N,D_S}) < r(pi_n(D_{N,D_S}))\big) f_D\big(D_{N,D_S}\big) \, \mathbf{d}D_S \mathbf{d}D_N \quad \text{(Equation 17)}$$

with f_D as the bivariate distribution of deposition.

4 Uncertainties in the RAINS Calculations of Ecosystems Protection

As a practical example, it was analyzed how uncertainties in important model terms propagate through the calculation chain of the RAINS model, i.e., from emissions over atmospheric dispersion to environmental impacts. For each of the elements, critical model terms were selected. An attempt was made to quantify the uncertainties of these model terms based on expert judgment.

The general concept of the analysis is outlined in Figure 3.1.

4.1 Uncertainties in Emission Estimates

The methodology adopted by the RAINS model to estimate emissions is associated with a variety of different types of uncertainties. Following the taxonomy introduced above, uncertainties are caused by the structure of the RAINS model. For instance, the selected aggregation level might influence calculation results, i.e., how emission sources are aggregated into certain economic sectors and categories of fuels. Uncertainties are also related to the need to determine mean values for a RAINS category that in reality encompasses an inhomogeneous range of emission sources. The determination of the expected values for fuel quality, emission control efficiencies and application rates of emission control measures causes important parameter uncertainties. For the forcing functions, uncertainty is an issue for the accuracy of statistical information on economic activities as well as for the projection of future activity rates. Depending on the operation mode of the RAINS model, the implementation of emission control measures (or compliance with legislation) could also be a major source of uncertainties. Uncertainties related to the initial state of the models are caused by estimates of the hypothetical uncontrolled emissions and the implementation of emission control measures control measures in the base year.

Model structure	Selected sectoral aggregation
	Determination of mean values
Parameters	Expected values for fuel quality, removal efficiencies and application
	rates
Forcing functions	Accuracy of statistical information on economic activities
	Projections of sectoral economic activities
	Future implementation of emission controls
Initial state	Uncontrolled emission factors
	State of emission controls in the base year

Table 4.1: Sources of uncertainties for the emission calculations in the RAINS model

4.1.1 Assumed Uncertainties in Input Data for SO₂ and NO_x

An attempt was made to quantify the uncertainties of some of the key model terms. Based on expert judgment, coefficients of variation (CV) were compiled for

- activity rates (*act*); in this case different uncertainties were specified for the base year 1990 and for the values projected for 2010,
- uncontrolled emission factors (*ef*) (mean values for a source category),
- efficiencies (rem) of the various emission control measures considered in the RAINS model, and
- the rates of application (*af*) of such measures.

For quantifying the uncertainties, four groups of countries are distinguished:

- The best data quality is assumed for EU-15 and EFTA countries (Norway, Switzerland), for which harmonized and quality controlled international statistics are available.
- The second group of countries includes the 'first wave' EU accession countries with a fairly advanced economic and administrational reform (the Czech Republic, Estonia, Hungary, Poland, Slovakia, Slovenia).
- All other countries are included in the third group.
- Little solid information is available for international sea traffic.

Uncertainties in Activity Data

Uncertainties in activity data originate from

- inaccuracies in measuring physical quantities of consumed fuels or manufactured products,
- errors and biases in measuring the heat content of fuels,
- possible international inconsistencies in fuel- and sector definitions of national statistics,
- the assumptions about future economic development and structural changes in energy systems, etc.

Although the RAINS database uses to the maximum possible extent internationally harmonized data sets with coherent definitions and compilation methods, reporting practices of countries are not always fully consistent. For instance, national statistics of different countries classify heating oil of a similar quality either as "heavy fuel oil" or as "light fuel oil". Also fuel consumption of industrial power plants and municipal heating plants is sometimes reported in the power plant sector, while in other cases it is included in the industrial or domestic sector, depending on the conventions used in individual countries.

For the base year 1990, the economic transition of the former centrally planned economies and the formation of new countries are additional sources of uncertainties, since new statistical systems for new economic agents (private firms) and new administrative units had to be created. Obviously, for such transition countries future projections of economic development are more uncertain than for countries with stable market economies.

Since SO_2 and NO_x calculations are based on the same activity data, errors introduced by this term into SO_2 and NO_x calculations are perfectly correlated.

	1990	2010
EU-15, Norway and Switzerland	0.06/0.10	0.12/0.18
Czech Rep., Estonia, Hungary, Poland, Slovakia, Slovenia	0.09/0.15	0.15/0.22
Other countries	0.12/0.18	0.18/0.27
International sea traffic	0.20	0.30
	EU-15, Norway and Switzerland Czech Rep., Estonia, Hungary, Poland, Slovakia, Slovenia Other countries International sea traffic	1990EU-15, Norway and Switzerland0.06/0.10Czech Rep., Estonia, Hungary, Poland, Slovakia, Slovenia0.09/0.15Other countries0.12/0.18International sea traffic0.20

Table 4.2: Groups of countries/regions for which different assumptions on uncertainties in emission data were made, and the coefficients of variation (CV) for the activity data (sectoral fuel consumption, industrial production)

Uncertainties in Emission Factors

Many factors contribute to uncertainties of the uncontrolled emission factors that are used to calculate SO_2 and NO_x emissions in the RAINS model. For instance, the extrapolation of emission factors that were monitored for a few individual sources under certain conditions to entire sectors, different operating conditions and other countries is certainly an important aspect. It is well known that emissions often depend on the age of the equipment and on maintenance, and it is difficult for many reasons to accurately reflect this in emission inventories.

Source-group specific uncertainties in the uncontrolled emission factors are assumed for this analysis (Table 4.3 for SO₂, Table 4.4 for NO_x). For SO₂ emission factors of Group I countries, a CV of 0.05 has been adopted for most sectors. Larger uncertainties are associated with SO₂ and NO_x emissions from brown coal combustion due to the greater variability of fuel quality even from the same coalmine and the limited possibility to stabilize combustion conditions. Higher uncertainties prevail also in the transport sector, where the determination of emission factors that are representative for the entire vehicle fleet, driving conditions and maintenance level is difficult.

To reflect conditions where less effort is spent to determine country-specific emission factors, where lower levels of maintenance prevail or where fast changes in technology are expected, the CVs given in Table 4.3 are increased by 25 percent for Group II and 50 percent for Group III countries. For international sea traffic 50 percent larger CVs are assumed.

Generally higher uncertainties are assumed for NO_x, emission factors.

No correlations between the uncertainties of emission factors of different source categories are assumed.

	Brown coal	Hard coal	Coke, briquettes	Gas	Heavy fuel oil	Diesel, light fuel oil	Gasoline	Other solids
Refineries, coke prod.	0.10	0.05	0.05	0.05	0.05	0.05	0.05	
Power plants	0.10	0.05	0.05	0.05	0.05	0.05		0.05
Industry	0.10	0.05	0.05	0.05	0.05	0.05		0.05
Domestic	0.10	0.05	0.05	0.05	0.05	0.05		0.05
Transport					0.05	0.05	0.05	
Industrial process emissions				0.	10			

Table 4.3: Coefficients of variation for uncontrolled SO_2 emission factors for Group I (EU and EFTA) countries. Values for Group II countries are increased by 25 percent, for Group III countries by 50 percent.

Table 4.4: Coefficients of variation for uncontrolled NO_x emission factors for Group I (EU and EFTA) countries. Values for Group II countries are increased by 25 percent, for Group III countries by 50 percent.

	Brown coal	Hard coal	Coke, briquettes	Gas	Heavy fuel oil	Diesel, light fuel oil	Gasoline	Other solids
Refineries, coke prod.	0.15	0.075	0.075	0.075	0.075	0.075	0.075	
Power plants	0.15	0.075	0.075	0.075	0.075	0.075		0.075
Industry	0.15	0.075	0.075	0.075	0.075	0.075		0.075
Domestic	0.15	0.075	0.075	0.075	0.075	0.075		0.075
Road transport					0.075	0.1125	0.1125	
Off-road transport					0.075	0.1125	0.1875	
Industrial process emissions				0.1	15			

Uncertainties in Removal Efficiencies

Without any doubt there are also significant uncertainties associated with determining mean removal efficiencies of the emission control measures in a country. For instance, design efficiencies might vary over Europe, local fuel quality may influence the performance of emission abatement devices, and plant operators have certain freedom to run their equipment in a more or less efficient way.

For this analysis it is assumed that for a number of reasons uncertainties in the efficiency of technically more advanced techniques are lower than those of simple measures. In many cases

advanced techniques (e.g., high efficiency flue gas desulfurization) are applied in plants with continuous monitoring of emissions, where compliance with stringent emission standards is required. Also if standards for fuel quality are very tight (e.g., sulfur content of diesel oil), controls are often more stringent.

Coefficients of variation (CV) for SO_2 and NO_x control technologies are presented in Table 4.5 and Table 4.6. These coefficients are uniform for all countries in Europe.

Table 4.5: Assumed uncertainties of the removal efficiency parameter used in the SO_2 emission calculations (Equation 4). The uncertainty is presented as the coefficient of variation.

Emission control options	Coefficient of variation (CV) for removal efficiency
Low sulfur fuels for stationary sources (coal, coke, heavy fuel oil, gas oil)	0.05
Low sulfur diesel for transport sources	0.005
Limestone injection, fluidized bed combustion	0.04
High efficiency flue gas desulfurization	0.005
Industrial process emissions - Stage 1	0.05
Industrial process emissions - Stage 2	0.035
Industrial process emissions - Stage 3	0.03

Table 4.6: Assumed uncertainties of the removal efficiency parameter used in the NO_x emission calculations (Equation 4). The uncertainty is presented as coefficient of variation.

Emission control options	Coefficient of variation (CV) for removal efficiency
Combustion modification for stationary sources	0.05
SCR/SNCR for stationary sources (also in combination with combustion modification)	0.05
EURO 1 standards for gasoline vehicles	0.075
EURO 2 standards for gasoline vehicles	0.05
EURO 3 standards for gasoline vehicles	0.025
EURO 4 standards for gasoline vehicles	0.015
EURO 1 and 2 standards for diesel vehicles	0.075
EURO 3 standards for diesel vehicles	0.05
EURO 4 standards for diesel vehicles	0.025
Control of industrial process emissions - Stage 1	0.10
Control of industrial process emissions - Stage 2	0.075
Control of industrial process emissions - Stage 3	0.05

4.1.2 Assumed Uncertainties in Input Data for NH₃

The calculations of ammonia emissions in the RAINS model follows a slightly different path, since the several phases during which emissions could occur are treated separately. Thereby it is possible to associate differentiated uncertainties to the various stages. To reflect differences in the quality of available information for western European and central and eastern European countries, two groups of countries are considered.

Uncertainties in Activity Data

General uncertainty estimates for animal numbers (i.e., number of dairy cows, fattening pigs, laying hens, et.) and fertilizer use statistics (SAEFL, 2001) are not available. Van der Hoek (1995) suggests that, e.g., for the Netherlands, the uncertainty of animal number statistics is in the range of 10 percent. Obviously uncertainties will increase for future projections. Assumptions for this study are listed in Table 4.7.

	EU-15 + EFTA		Other co	ountries
	1990	2010	1990	2010
Cattle	0.075	0.15	0.10	0.20
Pigs	0.075	0.15	0.10	0.20
Poultry	0.10	0.20	0.15	0.30
Fur animals	0.05	0.10	0.05	0.10
Horses	0.05	0.10	0.10	0.20
Sheep	0.15	0.30	0.15	0.30
Application of other N fertilizers	0.05	0.10	0.10	0.20
Application of urea	0.05	0.10	0.10	0.20
N fertilizer production	0.05	0.10	0.075	0.15
Other industry	0.05	0.10	0.15	0.30
Waste treatment	0.15	0.30	0.30	0.60
Other sources	0.25	0.50	0.50	1.00

Table 4.7: Uncertainties in the activity data for ammonia, coefficients of variation

Uncertainties in Emission Factors

In general there is good understanding of the processes that cause ammonia emissions, with the possible exception for emissions from solid manure. However, there are still large uncertainties in the determination of the parameters that are needed to derive country specific emission factors. Since actual measurement studies were conducted only in a few countries (mainly in western Europe), brave assumptions have to be made for extrapolating results of such studies to other countries with sometimes rather different conditions.

Van der Hoek (1995) reports a typical uncertainty range of 30 percent for livestock emission factors, not considering the inter-annual meteorological differences that might greatly affect actual ammonia losses from manure management.

Sutton (1995) indicates that for emission factors for fertilizer application the major source of uncertainties is the generalization of emission factors rather than the area of crops under cultivation. Generally he estimates an uncertainty of about 50 percent.

Several measurement campaigns indicated the importance of fertilizer application practice on ammonia losses (e.g., Isherwood, 2001; UN/ECE, 1999), especially for urea fertilizers. However, there is an important lack of detailed information on national practices to handle manure and fertilizers. A coefficient of variation of 40 percent is assumed in this study for the urea emission factor, while for other N-fertilizers the CV is assumed to be 25 percent (Table 4.8).

	Housing	Storage	Manure application	Grazing	Non animal
Cattle	0.075	0.15	0.20	0.20	
Pigs	0.15	0.20	0.20		
Poultry	0.15	0.15	0.20		
Fur animals	0.25	0.35	0.20	0.25	
Horses	0.25	0.35	0.20	0.25	
Sheep	0.25	0.35	0.20	0.25	
Application of other N fertilizers					0.25
Application of urea					0.40
N fertilizer production					0.15
Other industry					
Waste treatment					
Other sources					

Table 4.8: Uncertainties of emission factors used for the calculation of ammonia emissions for western European countries. The uncertainty is presented as coefficient of variation (CV).

Table 4.9: Uncertainties for emission factors used for the calculation of ammonia emissions for central and eastern European countries. The uncertainty is presented as coefficient of variation (CV).

Animal category	Housing	Storage	Manure	Grazing	Non
Ammar category	Housing	Storage	application	Orazing	animal
Cattle	0.20	0.30	0.30	0.40	
Pigs	0.20	0.30	0.30		
Poultry	0.20	0.30	0.30		
Fur animals	0.25	0.35	0.30	0.25	
Horses	0.25	0.35	0.30	0.25	
Sheep	0.25	0.35	0.30	0.25	
Application of other N fertilizers					0.25
Application of urea					0.40
N fertilizer production					0.15
Other industry					
Waste treatment					
Other sources					

Uncertainties in the Reduction Efficiencies of Emission Control Techniques

The aggregation of various emission sources into categories for which several emission control options with varying efficiencies are available constitutes an important source of uncertainty when determining the reduction efficiency. Furthermore, it was shown that even the same techniques perform differently depending on several parameters such as soil types, application practice, etc. Several studies explored the ranges of efficiencies for various technologies (UN/ECE, 1999; Webb, 2001). For this study we assume that the uncertainty of reduction efficiencies ranges typically between 20 and 40 percent. For some techniques, e.g., housing adaptation in Eastern Europe, the uncertainty could reach up to 80 percent owing to poor information on current practices.

Since the RAINS methodology calculates emissions through a chain of different states, correlations between reduction efficiencies at the different stages are important.

		Low NH ₃		Low nit	rogen feed		Housing a	daptation
	Covered storage	manure application	Housing	Storage	Manure spreading	Grazing	Housing	Storage
Cattle, liquid manure	0.05	0.1	0.2	0.2	0.2	0.15	0.2	0.05
Cattle, solid manure		0.15	0.2	0.2	0.2	0.15		
Pigs, liquid manure	0.05	0.1	0.2	0.2	0.2	0.15	0.1	0.05
Pigs, solid manure		0.15	0.2	0.2	0.2	0.15		
Laying hens	0.05	0.1	0.2	0.2	0.2	0.15	0.075	0.05
Other poultry	0.05	0.1	0.2	0.2	0.2	0.15	0.075	0.05
Sheep		0.1						

Table 4.10: Uncertainty in the removal efficiencies. The uncertainty is presented as the coefficient of variation (CV)

4.1.3 Resulting Uncertainties in Emission Calculations

With the uncertainties of input data as specified in the preceding section and equations 3 and 4, uncertainties of the resulting emission estimates can be derived.

To present uncertainties of calculation results, we provide the standard deviation in absolute terms (kilotons) and the 95 percent confidence interval expressed as a percentage of the expected value. The confidence interval is calculated based on the assumption of a normal distribution of the overall uncertainty. (For normal distribution, the 95 percent confidence interval is roughly twice the standard deviation).

Emission uncertainties are evaluated on a sectoral basis for three countries (UK, Switzerland and Romania). The uncertainties of national total emissions are provided for all countries, and a sensitivity analysis that explores how much the individual factors contribute to the overall uncertainties is presented.

Sectoral Emission Estimates

Uncertainties of sectoral emission estimates are presented for two countries (UK and Switzerland) for 1990 and 2010.

For the UK, the assumptions described in the preceding section lead for the year 1990 to similar uncertainty ranges for the 10 most important source sectors (Table 4.11). An exception is the industrial process emission sector, where the emission factors are associated with larger uncertainties. Since we do not assume correlation between the parameters of different source sectors, errors compensate to a certain extent so that the overall uncertainty in national total emissions is smaller than that of individual sectors.

This situation looks different if we consider the projection for the year 2010 (Table 4.12). While the expected value of emissions declines by 66 percent, uncertainties increase mainly due to three reasons:

- First, we assume that our knowledge about future activity data is less certain than for the past.
- Second, the introduction of emission control devices adds an additional source of uncertainty to the emission estimates (the removal efficiency). For instance, as a consequence of the more uncertain activity data projections and the more stringent emission controls, the relative uncertainty range (the confidence interval) of emissions from coal power plants in the UK will increase from ±16 to ±26 percent. In

absolute terms, however, the uncertainty will certainly decline, e.g., the standard deviation from 199 kilotons in 1990 to 49 kilotons in 2010.

• Third, since many of the sectors that made major contributions in 1990 will reduce their emissions to a significant extent, other sectors that were in 1990 of minor relevance but associated with larger uncertainties will become relatively more important sources (e.g., emissions from aircraft and other sources). This effect will be exacerbated by the general trend to replace sulfur containing fuels such as coal and oil by natural gas.

These effects, which occur throughout the full range of emission sources, lead to an increase in the overall uncertainty for SO_2 emissions in the UK from 11 percent to 15 percent.

A similar picture emerges for NO_x emissions, where the expected value declines by 58 percent (Table 4.13, Table 4.14). Passenger gasoline vehicles, which made the largest contributions in 1990, will move out from the list of the 'top 10' sources, and emissions from diesel trucks will step up to the leading position. Overall uncertainties for the year 2010 are slightly larger than in 1990, mainly due to the uncertainties in projected activity levels.

It is interesting to note, however, that the overall uncertainties in the estimates for national total NO_x emissions is similar, or even slightly smaller, than the uncertainties for SO₂, which might be in contrast to many expert guesses. Indeed, this result is surprising at first glance, since in this calculation the uncertainty of the NO_x emission factors was consistently assumed to be between 50 to 350 percent higher than those for SO₂ – in line with general expert judgment. Uncertainties in the activity rates are obviously identical for SO₂ and NO_x, and the uncertainties of removal rates are similar. As a consequence, as to be expected, uncertainties of the estimates *for individual sectors* are higher for NO_x than for SO₂.

That the estimates of *national total emissions* in the UK are more certain for NO_x than for SO_2 is caused by the fact that SO_2 emissions are dominated (both in 1990 and 2010) by a few source sectors (i.e., power stations burning hard coal, responsible for 67 percent of SO_2 emissions in 1990 and for 39 percent in 2010, and industrial processes). This dominance of a few sectors restricts the potential for compensation of errors for the SO_2 calculations. Since NO_x emissions originate from several sectors with almost equal contributions, compensation of errors is a powerful mechanism leading to lower uncertainties in national total emissions.

It must be stressed that this effect is intrinsically linked to the selected aggregation and only occurs for national total emissions where a substantial potential for error compensation exists. Such a potential might not exist if smaller units are considered, e.g., geographical grid cells or local administrative districts.

It is also clear that this finding depends strongly on the composition of national emission sources, i.e., whether one or two sectors dominate total emissions in a country or not. This is illustrated by the case of Switzerland (Table 4.15 - Table 4.18), where for the year 1990, in the absence of dominating SO_2 emissions from coal power stations, the uncertainty is estimated at 9 percent for SO_2 and at 13 percent for NO_x emissions.

Rank	Fuel	Sector	SO ₂ emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Hard coal	Power plants, existing	2550	199	±16 %
2	Heavy fuel oil	Power plans, existing	350	27	±16 %
3		Industrial processes	214	30	±28 %
4	Heavy fuel oil	Refineries	120	9	±16 %
5	Hard coal	Domestic	103	8	±16 %
6	Heavy fuel oil	Industry	89	7	±16 %
7	Hard coal	Industry	82	6	±16 %
8	Diesel	National sea traffic	48	4	±16 %
9	Fuel oil	Domestic	43	3	±16 %
10	Diesel	Road transport	38	3	±16 %
				•••	
NATI	ONAL TOTAL	EMISSIONS	3812	204	±11 %

Table 4.11: SO_2 emissions and their uncertainties of the 10 largest emission source sectors in the United Kingdom in the year 1990.

Table 4.12: SO_2 emissions and their uncertainties of the 10 largest emission source sector in the United Kingdom for the year 2010.

D 1	T 1	Sector	SO ₂ emissions	Standard	95 percent
Rank	Fuel	Sector	(expected value	deviation	confidence
			[kt])	(kt)	interval
1	Hard coal	Power plants, existing	379	49	±26 %
2		Industrial processes	216	49	±45 %
3	Heavy fuel oil	Power plants, existing	145	23	±32 %
4	Diesel	National sea traffic	43	6	±26 %
5	Heavy fuel oil	Refineries	29	4	±31 %
6		Air traffic and other uncontrollable sources	27	6	±45 %
7	Heavy fuel oil	Domestic	23	4	±31 %
8	Hard coal	Industry	17	2	±26 %
9	Heavy fuel oil	International sea traffic	17	2	±26 %
10	Hard coal	Domestic	15	2	±26 %
NATI	ONAL TOTAL	EMISSIONS	962	74	±15 %

Rank	Fuel	Sector	NO _x emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Gasoline	Passenger cars	781	100	±26 %
2	Hard coal	Power plants, existing	696	67	± 19 %
3	Diesel	Heavy duty trucks	506	65	±26 %
4	Diesel	Off-road machinery	144	18	±26 %
5		Industrial processes	129	23	±36 %
6	Heavy fuel oil	Power plants, existing	69	7	±19 %
7	Diesel	National sea traffic, large vessels	65	8	±26 %
8	Natural gas	Domestic	64	6	±19 %
9	Diesel	National sea traffic, medium vessels	60	8	±26 %
10		Air traffic, other uncontrollable sources	60	11	±36 %
•••					
]	NATIONAL TO	TAL EMISSIONS	2839	141	±10 %

Table 4.13: NO_x emissions and their uncertainties of the 10 largest emission source sectors in the United Kingdom, 1990.

Table 4.14: NO_x emissions and their uncertainties of the 10 largest emission source sectors in the United Kingdom for the year 2010.

D 1		Sector	NO _x emissions	Standard	95 percent
Rank	Fuel	Sector	(expected value [kt])	deviation (kt)	interval
1	D'1	II	200	26	× 25.0/
1	Diesel	Heavy duty trucks	208	36	$\pm 35\%$
2	Hard coal	Power plants, existing	171	25	±29 %
3		Industrial processes	134	34	±50 %
4	Diesel	Off-road transport	88	15	±33 %
5		Air traffic, other uncontrollable sources	86	22	±50 %
6	Natural gas	Domestic	84	12	±28 %
7	Natural gas	Power plants, new	59	9	±29 %
8	Diesel	National sea traffic, large vessels	59	10	±33 %
9	Diesel	National sea traffic, medium vessels	55	9	±33 %
10	Heavy fuel oil	Power plants, existing	49	7	±29 %
NATIO	NAL TOTAL E	EMISSIONS	1198	66	±11 %

Rank	Fuel	Sector	SO ₂ emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Fuel oil	Domestic	21.6	1.8	±16 %
2		Industrial processes	5.8	0.8	±29 %
3	Diesel	Heavy duty trucks	3.1	0.3	±16 %
4	Hard coal	Industry	2.5	0.2	±16 %
5	Heavy fuel oil	Power stations	1.9	0.2	±19 %
6	Fuel oil	Industry	1.8	0.2	±16 %
7	Diesel	Off-road machinery	1.5	0.1	±16 %
8	Heavy fuel oil	Refineries	1.0	0.1	±16 %
9	Diesel	Light duty vehicles	0.9	0.01	±16 %
10	Hard coal	Power stations	0.7	0.1	±16 %
NATI	ONAL TOTAL	EMISSIONS	43	2.0	±9 %

Table 4.15: SO_2 emissions and their uncertainties of the 10 largest emission source sector in Switzerland, 1990.

Table 4.16: SO_2 emissions and their uncertainties of the 10 largest emission source sector in Switzerland for the year 2010.

Rank	Fuel	Sector	SO ₂ emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Fuel oil	Domestic	8.6	1.1	±26 %
2		Industrial processes	4.2	1.0	±46 %
3	Hard coal	Industry	3.7	0.5	±26 %
4	Heavy fuel oil	Industry	1.9	0.3	±27 %
5	Heavy fuel oil	Refineries	1.8	0.2	±26 %
6	Light fuel oil	Industry	0.9	0.1	±26 %
7	Wood, waste	Domestic	0.8	0.1	±31 %
8	Waste	Industry	0.8	0.1	±31 %
9	Heavy fuel oil	Power stations	0.8	0.1	±28 %
10	Other	Air traffic, waste treat.	0.5	0.1	±45 %
NATI	IONAL TOTAL	EMISSIONS	26	1.6	±13 %

Rank	Fuel	Sector	NO _x emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Gasoline	Passenger cars	72.4	9.7	±27 %
2	Diesel	Heavy duty trucks	26.8	3.6	±27 %
3	Diesel	Off-road machinery	15.4	2.0	±26 %
4	Fuel oil	Domestic	13.7	1.3	±19 %
5		Industrial processes	10.0	1.9	±37 %
6	Other	Air traffic, waste, etc.	6.6	1.2	±36 %
7	Diesel	Light duty vehicles	3.0	0.4	±26 %
8	Gasoline	Off-road machinery	2.2	0.4	±39 %
9	Waste	Heat&power generation	2.1	0.3	±32 %
10	Natural gas	Domestic	2.0	0.2	±19 %
NATI	ONAL TOTAI	LEMISSIONS	163	10.8	±13 %

Table 4.17: NO_x emissions and their uncertainties of the 10 largest emission source sectors in Switzerland, 1990.

Table 4.18: NO_x emissions and their uncertainties of the 10 largest emission source sectors in Switzerland for the year 2010.

Rank	Fuel	Sector	NO _x emissions (expected value [kt])	Standard deviation (kt)	95 percent confidence interval
1	Diesel	Heavy duty trucks	21.0	3.6	±35 %
2	Fuel oil	Domestic	10.9	1.6	±28 %
3	Diesel	Off-road machinery	9.5	1.6	±33 %
4	Gasoline	Passenger cars	6.7	1.8	±55 %
5		Air traffic, other uncontrollable sources	6.6	1.7	±50 %
6		Industrial processes	5.3	1.5	±55 %
7	Waste	Industry	2.9	0.6	±39 %
8	Natural gas	Domestic	2.6	0.4	±28 %
9	Natural gas	Industry	2.2	0.3	±29 %
10	Diesel	Light duty vehicles	2.0	0.4	±34 %
NATIO	NAL TOTAL H	EMISSIONS	79	5.2	±13 %

It is interesting to rank the uncertainties with the largest impacts on the final results. As displayed in Table 4.19 for the UK (and Table 4.20 for Switzerland), results are strongly dependent on the specific situation, i.e., the pollutant and the year. For SO_2 in the base year, almost equal uncertainties originate from the activity data and emission factors. If emissions are projected in the future, estimates will be heavily influenced by the uncertainty of projected economic activities. For NO_x , however, base year estimates are more sensitive towards emission factors, while the influence of activity data is lower than for SO_2 . NO_x projections depend more on the uncertainties of activity data, and also the removal efficiency is of importance.

Table 4.19: Results from a sensitivity analysis: 95 percent confidence intervals for estimates of national SO_2 and NO_x emissions in the UK. Values describe the 95 percent confidence interval in national total emissions if only the uncertainties of the particular parameter are considered and the uncertainties for all other parameters are excluded.

	S	O ₂	N	O _x
	1990	2010	1990	2010
Activity data	±8 %	±14 %	±5 %	±8 %
Emission factors	±7 %	±6 %	±9 %	±7 %
Removal efficiency	±0 %	±3 %	±0 %	±3 %
All factors considered	±11 %	±15 %	±10 %	±11 %

Table 4.20: Results from a sensitivity analysis: 95 percent confidence intervals for estimates of national SO_2 and NO_x emissions of Switzerland. Values describe the 95 percent confidence interval in national total emissions if only the uncertainties of the particular parameter are considered and the uncertainties for all other parameters are excluded.

	S	O ₂	N	O _x
	1990	2010	1990	2010
Activity data	±7 %	±11 %	±6 %	±9 %
Emission factors	±6 %	±5 %	±11 %	$\pm 8~\%$
Removal efficiency	±3 %	±2 %	±4 %	±5 %
All factors considered	±9 %	±13 %	±13 %	±13 %

Uncertainties in National Total Emissions

As a general feature, uncertainties in national total emissions are much smaller than the uncertainties of the estimates for individual sectors. This is a consequence of the assumption that the uncertainties of the individual parameters (except activity rates) are not correlated, so that errors can efficiently compensate each other.

However, there are large differences in the uncertainties of national total emissions of different countries (Table 4.21, Table 4.22). For the year 1990, the 95 percent confidence interval of SO₂ estimates ranges from ± 6 percent for France to ± 23 percent for the Former Yugoslavia. For NO_x estimates, the range spans from ± 8 percent for Greece to ± 23 percent for

the Former Yugoslavia, and for NH_3 from ± 9 percent for Sweden to ± 23 percent for Albania. Several factors contribute to larger uncertainties:

- If emissions in a country are dominated by one or two source sectors, there is only a limited potential that errors in the estimates of these sectors can be statistically compensated by other sectors (for instance, this is the case for SO₂ emissions in the UK).
- If parameters (activity data or emission factors) of a large sector are especially uncertain (e.g., lignite burning in the Czech Republic), this will have an influence on national total emissions.
- If many parameters are more uncertain, e.g., in central and eastern European countries.

Uncertainties of SO₂ estimates for the base year are generally between 10 and 15 percent, which is in line with what is expected by many emission inventory experts. More interesting, however, is the fact that, with the specific assumptions about the uncertainties of the input parameters, estimates of NO_x emissions are not significantly more uncertain than \hat{SO}_2 estimates, which somehow contradicts the general opinion of experts who often suggest (however not based on quantitative methods) for NOx estimates an "uncertainty" of ± 30 percent. Along these lines of arguments we have also assumed in this analysis larger uncertainties in the input data that are relevant for NO_x emissions (e.g., emission factors) and, based on the literature and expert judgment, we believe that the assumed uncertainties are not underestimated. Indeed, the sectoral analysis in the preceding section confirms that sectoral emission estimates for NO_x are clearly more uncertain than those for SO_2 (compare, e.g., Table 4.13 with Table 4.11). Still, the overall uncertainties in national total emissions for NO_x are not significantly higher than those of SO_2 . This is mainly caused by the fact that in the case of NO_x several sectors (passenger cars, heavy duty trucks, power stations, process emissions) make similar contributions to total emissions, while for SO_2 the emissions of a few source sectors (power stations) are often dominating and thereby limiting the potential for error compensation. The same mechanism applies to NH₃ emissions, so that also for this pollutant, despite all the large uncertainties in many of the input data, the 95 percent confidence interval for national total emissions ranges between ± 10 and ± 20 percent.

In most cases, future emissions are more uncertain than estimates for historical years, which is an obvious consequence of the larger uncertainty associated with the projections of activity data. There are, however, notable exceptions mainly in countries in economic transition where, due to the restructuring process, some of the above mentioned factors that led in the year 1990 to large uncertainties become less important, so that the overall uncertainties will decline. This is the case, e.g., for the Czech Republic, where reduced lignite consumption decreases the uncertainties of SO_2 estimates from 20 to 17 percent and that of NO_x estimates from 18 to 16 percent. A similar effect can be observed for the New German Länder.

It should be noted that estimates of emissions from international sea traffic are consistently associated with largest uncertainties, due to the significant uncertainties in many of the relevant input data and the homogeneity of the source sector, which provides only limited scope for compensation of statistically independent errors.

The above findings are to a large extent related to the assumption that errors in input data of the different source sectors are uncorrelated, with the exception of activity data, which are common input to SO_2 and NO_x estimates. Indeed there is little evidence that errors in emission factors and emission control efficiencies (especially for SO_2 and NO_x) would be correlated (e.g., across sectors), and if so, even the sign of a possible correlation could be questionable. For instance, there might be a connection between the sulfur content of coals used in power stations and households. Let's construct a case where the assumed sulfur content of coal used in the domestic sector is too high. There is no generic evidence whether in such a case the sulfur content would be too high in the power sector too (because the statistical information of coal quality in the country is wrong), or whether the high sulfur coal

is predominantly used in the power sector, so that the sulfur content of coal for electricity generation should be even higher, if the country average sulfur content is accurate. Similar arguments could be constructed for, e.g., the emission factors of ammonia, where the interdependencies of the various emission stages could lead to negative correlations.

Perfect correlation has been assumed for the activity data that are used to estimate SO_2 and NO_x emissions. Consequently, SO_2 and NO_x estimates are also correlated with each other, depending on the structure of emission sources in a country. In countries with comparably little SO_2 emissions from coal and oil the correlation is weak (e.g., Austria 0.03, Netherlands 0.05), while the correlation could reach a value of up to 0.27 in other cases (e.g., Poland). Most notably, largest correlations (0.29) occur for international sea traffic, which has heavy fuel oil as the only energy source. Correlations will increase in the future, when the importance of potential errors in emission factors will be reduced (due to emission controls), while the uncertainty of the activity data, for which the correlation is specified, will be higher.

	S	O ₂	N	O _x	SO_2/NO_x	N	H_3
	Expected value (kt)	95 percent confidence interval	Expected value (kt)	95 percent confidence interval	Correlation	Expected value (kt)	95 percent confidence interval
Albania	72	±10%	24	±12%	0.08	32	±23%
Atlantic Ocean	641	±19%	911	+26%	0.29	_	
Austria	93	+9%	192	+10%	0.03	77	+10%
Baltic Sea	72	+19%	80	+26%	0.29		_10/0
Belarus	843	+12%	402	+11%	0.16	219	+17%
Belgium	336	+13%	351	+13%	0.04	97	+11%
Bosnia-Herczeg	487	+19%	80	+15%	0.17	31	+16%
Bulgaria	1842	+21%	355	+13%	0.06	141	+18%
Croatia	180	± 2170 +10%	82	$\pm 13\%$	0.00	40	±10%
Czech Ren	1873	±10%	546	$\pm 14\%$	0.11	107	$\pm 10\%$
Denmark	187	$\pm 20\%$	274	±10%	0.11	77	±1 1 7%
Estonia	275	±10%	274	<u>+</u> 970	0.28	20	$\pm 12/0$ $\pm 170/$
Estonia	215	±18%	04 276	$\pm 13\%$	0.12	29 40	±1770 ±1004
Fillialla	1250	±0%	270	±9% ⊤110/	0.00	40 810	±10%
Commony Novi	1230	±0%	702	$\pm 11\%$	0.00	201	$\pm 160/$
Germany, NewL.	4438	±10%	102	$\pm 13\%$	0.18	201	$\pm 10\%$
Germany, Old L.	842 504	±0%	1900	$\pm 11\%$	0.07	220	±11%
Greece	504	±/%	345 210	±8%	0.10	80	±21%
Hungary	913	±16%	219	±12%	0.06	120	±18%
Ireland	1/8	±/%	113	±9%	0.21	127	$\pm 13\%$
Italy	1679	±11%	2037	±9%	0.10	462	±14%
Latvia	121	±8%	117	±11%	0.08	43	±16%
Lithuania	213	±12%	153	±11%	0.11	80	±16%
Luxembourg	14	$\pm 14\%$	22	±12%	0.02	7	±15%
FYRMacedonia	107	±22%	39	±22%	0.09	17	±17%
Mediterranean	12	$\pm 19\%$	13	±26%	0.29		
Rep. of Moldova	197	$\pm 10\%$	87	$\pm 10\%$	0.17	47	$\pm 14\%$
Netherlands	201	±10%	542	$\pm 9\%$	0.05	233	±13%
North Sea	439	$\pm 19\%$	639	±26%	0.29		
Norway	52	$\pm 17\%$	220	$\pm 11\%$	0.02	23	$\pm 14\%$
Poland	3001	$\pm 11\%$	1217	±12%	0.27	505	$\pm 17\%$
Portugal	343	$\pm 8\%$	303	±10%	0.14	77	$\pm 10\%$
Romania	1331	$\pm 17\%$	518	$\pm 11\%$	0.07	292	$\pm 15\%$
Russia Kalinin.	44	$\pm 11\%$	29	$\pm 11\%$	0.13	11	$\pm 14\%$
Russia, Kola-K.	739	$\pm 18\%$	111	±12%	0.06	6	$\pm 14\%$
Russia, Other	2021	1.80/	2126	+ 1 1 0/	0.06	1001	150/
Dussia St Datars	200	±0%	221	$\pm 11\%$	0.00	1221	$\pm 1.3\%$
Russia St. Feleis.	500	$\pm 12\%$	221	±11%	0.13	44 60	±14%
Siuvakia	34ð 200	±12%	219 20	$\pm 12\%$	0.09	00	エ19% 」100/
Sioveilla	200	±20%	0U 1160	±13%	0.09	25 250	±19%
Spain	2189	±12%	1102	±9%	0.00	552	±13%
Sweden	11/	±9%	558	±10%	0.05	61 72	±9%
Switzerland	43	±9%	163	±1.5%	0.08	12	±13%
Ukraine	3706	±9%	1888	±10%	0.15	/29	±15%
United Kingdom	3812	±11%	2839	±10%	0.23	329	±12%
Yugoslavia	585	±23%	211	±23%	0.09	90	±14%

Table 4.21: Expected emissions, emission uncertainties and correlation between SO_2 and NO_x emissions in 1990.

	SO_2		NO _x		SO ₂ /NO _x	NH ₃	
	Exported	95 percent	Expected	95 percent	Correlation	Expected	95 percent
	value (kt)	confidence	value	confidence	Correlation	value	confidence
	value (Kt)	interval	(kt)	interval		(kt)	interval
Albania	55	9%	36	22%	0.20	35	23%
Atlantic Ocean	641	28%	911	33%	0.62		
Austria	39	15%	97	12%	0.07	67	15%
Baltic Sea	72	28%	80	33%	0.62		
Belarus	494	14%	316	15%	0.20	163	17%
Belgium	171	24%	169	16%	0.02	96	17%
Bosnia-Herczeg.	415	19%	60	14%	0.23	23	15%
Bulgaria	846	22%	297	17%	0.06	126	20%
Croatia	70	15%	91	19%	0.04	37	22%
Czech Rep.	336	17%	312	16%	0.29	108	14%
Denmark	146	20%	141	10%	0.38	72	15%
Estonia	107	24%	49	16%	0.20	29	20%
Finland	137	17%	149	11%	0.13	31	13%
France	574	16%	860	12%	0.03	780	14%
Germany, NewL.	141	15%	219	12%	0.21	147	15%
Germany, Old L.	372	12%	868	12%	0.12	425	14%
Greece	508	13%	342	10%	0.16	74	33%
Hungary	227	28%	159	15%	0.06	137	23%
Ireland	119	15%	79	10%	0.56	130	18%
Italy	381	22%	1013	13%	0.06	432	17%
Latvia	71	10%	84	14%	0.16	35	22%
Lithuania	61	16%	95	17%	0.11	81	17%
Luxembourg	8	36%	10	17%	0.01	9	25%
FYRMacedonia	81	20%	29	18%	0.17	16	23%
Mediterranean	12	28%	13	33%	0.62		
Rep. of Moldova	117	11%	66	13%	0.23	48	19%
Netherlands	76	21%	247	12%	0.03	141	15%
North Sea	439	28%	639	33%	0.62		
Norway	32	30%	178	16%	0.06	21	18%
Poland	1453	15%	728	11%	0.35	541	14%
Portugal	195	15%	259	13%	0.18	73	16%
Romania	594	20%	458	13%	0.08	304	17%
Russia Kaliningr.	18	16%	25	18%	0.10	11	19%
Russia, Kola-K.	473	34%	86	14%	0.03	4	14%
Russia Other	1717	12%	2517	15%	0.06	845	14%
Russia St.Peters.	136	18%	170	14%	0.16	33	14%
Slovakia	137	13%	132	16%	0.06	47	19%
Slovenia	114	30%	57	19%	0.11	21	22%
Spain	1006	15%	849	11%	0.10	383	18%
Sweden	65	17%	189	12%	0.10	61	12%
Switzerland	26	13%	79	13%	0.19	66	20%
Ukraine	1506	13%	1433	13%	0.14	649	14%
United Kingdom	962	15%	1198	11%	0.22	297	17%
Yugoslavia	269	25%	152	18%	0.16	82	14%

Table 4.22: Expected emissions, emission uncertainties and correlation between SO_2 and NO_x emissions for the year 2010.

4.2 Uncertainties in the Atmospheric Dispersion Calculations

Any calculation of the dispersion of sulfur and nitrogen compounds is loaded with many different types of uncertainties (Table 4.23). Some of the uncertainties are inherent in the atmospheric dispersion model that is used to derive the reduced-form representations of the atmospheric transport of pollutants in RAINS, others are related to the way such reduced-form descriptions are used in the RAINS model.

Model structure	Linearity in atmospheric dispersion of sulfur and nitrogen compound				
	Selected spatial resolution, ignoring in-grid variability				
	Country size (country-to-grid)				
Parameters	Expected values of parameters for describing chemical and physical processes (conversion rates, deposition rates)				
	Inter-annual meteorological variability				
Forcing functions	Spatial distribution of emissions within countries				
	Accuracy of meteorological data				
Initial state	Natural emissions				
	Hemispheric background				

Table 4.23: Types of uncertainties in the atmospheric dispersion calculations

For practical reasons it was not possible for the authors to address uncertainties of the underlying dispersion model, i.e., the EMEP model on the long-range transport of sulfur and nitrogen compounds in Europe. Recognizing the need for further analysis on this subject, a pragmatic approach was taken instead and the analysis used estimates of the uncertainties introduced by the inter-annual meteorological variability as a surrogate to demonstrate the methodology.

4.2.1 Assumed Uncertainties in Input Data

In the calculations of the RAINS model, fields of sulfur and nitrogen deposition are estimated based on transfer matrices that describe the dispersion of pollutants for the meteorological conditions of an entire year. At the moment matrices are available for 11 years (1985-1995). Since the ecological impacts of acid deposition that are addressed in the RAINS model are caused by long-term accumulated deposition, RAINS calculations use mean transfer matrices that average the dispersion characteristics over these 11 years. Such '11 years mean matrices' are used to analyze the present situation as well as to assess future trends in acid deposition.

Leaving the uncertainties that are inherent in the EMEP model aside, an attempt was made to quantify the uncertainties of the mean transfer matrices that are caused by fact that data for only 11 years are available.

The inter-annual meteorological variability σ_{tc} of the transfer coefficients $tc_{x,i,e}$ can be estimated from the dispersion coefficients of individual years $tc_{x,i,e,t}$:

$$Cov[tc_{x,i,e}, tc_{y,i,e}] = \frac{\sum_{t} (tc_{x,i,e,t} - tc_{y,i,e})(tc_{y,i,e,t} - tc_{y,i,e})}{n-1}$$
(Equation 18)

The variance of the transfer coefficients σ_{tc}^2 are the covariances of the transfer coefficient itself for the 11 (*n*) years.

Unfortunately the available 11 years data do not give us a fully representative picture of the long-term conditions. We use the central limit theorem (e.g., Milton & Arnold, 1995) to extrapolate the standard deviation σ_{tc} of the transfer coefficients computed for the mean of these 11 years data to long-term conditions $\sigma_{\mu tc}$:

$$\sigma_{\mu_{ic}} = \frac{\sigma_{tc}}{\sqrt{n}}$$
 (Equation 19)

The central limits theorem also allows us to derive the correlation of the transfer coefficients between the pollutants of a given emitter source, since this correlation is identical with the inter-annual correlation (Schönfeld, 1971):

$$\rho_{tc_{x,i,e},tc_{y,i,e}} = \frac{Cov[tc_{x,i,e},tc_{y,i,e}]}{\sigma_{tc_{x,i,e}}\sigma_{tc_{y,i,e}}}$$
(Equation 20)

4.2.2 Resulting Uncertainties in Atmospheric Dispersion Calculations

The combined uncertainties in deposition resulting from the uncertainties of the emission and atmospheric dispersion calculations are displayed in Figure 4.1 for 1990 and Figure 4.2 for 2010. These maps show for each grid cell the relative uncertainties (i.e., the confidence intervals divided by the expected values) of the deposition fields of sulfur, oxidized nitrogen, reduced nitrogen (from ammonia emissions) and total (oxidized and reduced) nitrogen.

In the year 1990 estimates of sulfur deposition are most uncertain in the new Länder of Germany, the Kola-Karelia region and in Romania with relative uncertainties in the range between ± 20 and ± 25 percent. These large uncertainties can be traced back to (i) relatively high uncertainties in the estimates of national emissions, (ii) the uncertainties introduced by the inter-annual meteorological variability, and (iii) situations where deposition at a given site is dominated by a few emission sources (countries), so that there is only a limited potential for compensation of errors. Especially the last cause applies to Kola-Karelia, where sulfur deposition originates to a large extent from the few smelters in the region, and also to Romania, where transboundary contributions do play a smaller role than in other countries.

In general, deposition estimates for nitrogen oxides appear more certain than those for sulfur, partly caused by the fact that uncertainties in national NO_x emissions are smaller than those for SO₂. Another reason is that oxidized nitrogen has longer travel distances in the atmosphere, so that the dispersion characteristics are 'better mixed' at least for long-term periods and that contributions to deposition from individual countries are more evenly distributed. This increases the potential for error compensation and reduces the uncertainties in estimates of total deposition. Reduced nitrogen, on the other hand, has shorter travel distances with more local dispersion characteristics, which results in comparably larger uncertainties of deposition estimates. Most interesting, however, is the fact that estimates of deposition of total nitrogen, i.e., of oxidized and reduced nitrogen compounds, is more certain than those for the individual compounds, generally within a range of ± 10 percent. This is a

consequence (i) of the larger number of sources, which increases the compensation potential and (ii) the negative correlation in the inter-annual meteorological variability of the transfer coefficients. This negative correlation is explained by the influence of meteorology on the short-term chemical reactions of nitrogen compounds: Sunny and warm weather increases the conversion from NO to NO_3^- , which shortens the travel distance of nitrogen oxides and increases deposition. Ammonia deposition is enhanced by wet weather conditions.

For the year 2010, uncertainties in deposition do not significantly change compared to 1990 in most cases. Exceptions are those countries which implement less than average emission reductions, so that their domestic contributions to total deposition will increase, which leads to lower potential for error compensation. The uncertainties in ammonia deposition will become a more important factor for total nitrogen deposition.



Deposition of total nitrogen

Deposition of reduced nitrogen

Figure 4.1: Ranges of relative uncertainties (confidence interval divided by the expected value) of the deposition estimates for the year 1990.



Deposition of total nitrogen



Figure 4.2: Ranges of relative uncertainties (confidence interval divided by the expected value) of the deposition estimates for the year 2010.

4.3 Uncertainties in Ecosystems Protection

4.3.1 Assumed Uncertainties in Input Data

Critical loads are derived from model calculations involving a large number of input parameters, all of them associated with significant uncertainties. Based on data for forest soils in Germany, the Coordination Center for Effects conducted a Monte Carlo analysis to determine the influence of uncertainties of input parameters on the quantification of protection isolines (Posch *et al.*, 2001). In particular, the analysis addressed uncertainties in

- base cation deposition,
- base cation uptake,
- base cation weathering,
- water flux from the bottom of the rooting zone,
- critical aluminum-base cation ratios,
- nitrogen immobilization,
- nitrogen uptake, and
- denitrification.

The analysis assumed parameters uniformly distributed around their median value and uncorrelated with the exception of base cation uptake and nitrogen uptake, for which full correlation was assumed.

Under the assumption that uncertainties in the critical loads estimates for different ecosystems are not correlated with each other, the uncertainty estimates derived for individual ecosystems also reflect the uncertainties of the protection isolines, because the relative order of ecosystems will not change when they are ranked according to their sensitivity.

For this analysis, the numerical values of the coefficients of variations as calculated by Posch *et al.* (2001), i.e., a CV of 0.22 for $CL_{max}(S)$ and a CV of 0.12 for $CL_{max}(N)$, are used as relative uncertainties along the isolines for all critical loads estimates.

4.3.2 Resulting Uncertainties in Ecosystems Protection

The uncertainties of ecosystems protection from emission scenarios are displayed in Figure 4.3 for 1990 and Figure 4.4 for 2010. Four maps are presented: (i) the ecosystems that are protected with 50 percent probability (similar to the deterministic case), (ii) the ecosystems that are protected with 95 percent probability, (iii) the ecosystems with 5 percent probability and (iv) the range of the 95 percent confidence interval in terms of percent ecosystems.

Uncertainties in ecosystems protection are determined by the uncertainties of the deposition estimates and the uncertainties of protection isolines. While we found that error compensation is one of the most important mechanisms determining uncertainties of emission and deposition estimates, for ecosystems protection the binary nature of the protection criterion (i.e., protected or not protected) becomes a dominating factor at places where deposition is close to critical loads or where critical load values for ecosystems in a grid cell are very similar.

This can be seen for 1990 in France and Russia, where the uncertainty ranges are largest because deposition was close to the critical loads and only few critical load values were reported for each grid cell.



Deterministic case, i.e., ecosystems protected with 50 percent probability



Ecosystems protected with 95 percent probability



Figure 4.3: Estimates of the percentage of protected ecosystems in the year 1990.



Deterministic case, i.e., ecosystems protected with 50 percent probability



Ecosystems protected with 95 percent probability



Figure 4.4: Estimates of the percentage of protected ecosystems in the year 2010.

In 2010, the expected value of ecosystems protection shows large improvement. However, uncertainty ranges are still significant and extend in many areas from 25 to 100 percent of the ecosystems.

The Gothenburg Protocol and the analyses for the EU Emission Ceilings Directive focused on the median estimate, for which there is a 50 percent probability that the environmental targets will be achieved and a 50 percent probability that the targets will be missed.

Table 4.24 shows for the year 2010 the ecosystems area that remains unprotected with certain probability. While with 50 percent probability 2.7 percent of Europe's ecosystems will remain unprotected (and 4.1 percent in the EU-15), there is a five percent probability ('the worst case') that in 2010 in Europe more than 7.2 percent (and 12.4 percent in the EU) remain unprotected. On the other hand, there is also a five percent chance (the 'best case') that only less than 0.9 percent (and 1.8 percent in the EU-15) remain unprotected leaving only few areas in Europe (at the Dutch/German border and in southern Norway) with excess deposition at more than 50 percent of the ecosystems area.

Thus, in the worst case (with five percent probability), Europe will massively fail to reach the envisaged environmental targets of the Gothenburg Protocol.

Table 4.24: Expected areas of ecosystems within grids in the region that remain unprotected in the year 2010 with different levels of confidence. The median (50%) estimate was used for negotiations of the Gothenburg Protocol.

	Probability								
	5%	10%	25%	50%	75%	90%	95%		
Austria	0.7%	1.2%	1.4%	2.7%	3.6%	9.1%	13.0%		
Belgium	11.1%	12.1%	15.6%	21.7%	24.5%	35.1%	37.4%		
Denmark	0.7%	0.8%	1.2%	2.6%	3.1%	4.3%	5.0%		
Finland	1.1%	1.3%	2.6%	3.8%	4.6%	6.3%	7.5%		
France	0.2%	0.3%	0.3%	0.7%	1.6%	7.7%	16.2%		
Germany	4.8%	6.3%	9.0%	13.5%	18.6%	29.6%	37.3%		
Greece	0.0%	0.0%	0.0%	0.0%	0.0%	0.2%	0.2%		
Ireland	0.4%	0.4%	0.8%	1.8%	3.7%	9.4%	12.2%		
Italy	1.5%	1.5%	1.9%	2.1%	3.0%	4.5%	6.2%		
Luxembourg	0.0%	1.0%	5.0%	6.0%	8.0%	13.0%	14.0%		
Netherlands	30.7%	36.7%	48.1%	59.1%	65.3%	74.5%	79.4%		
Portugal	0.0%	0.0%	0.0%	0.2%	0.2%	0.2%	0.2%		
Spain	0.1%	0.2%	0.2%	0.3%	0.3%	0.4%	0.5%		
Sweden	2.0%	2.5%	3.0%	4.1%	5.0%	7.0%	8.1%		
UK	6.3%	7.3%	9.3%	12.2%	15.4%	21.3%	24.2%		
EU-15	1.8%	2.1%	2.9%	4.1%	5.4%	9.1%	12.4%		
Albania	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%		
Belarus	2.9%	6.3%	10.4%	17.3%	22.1%	29.0%	31.4%		
Bosnia-H	0.0%	0.0%	0.0%	0.0%	9.1%	9.1%	9.1%		
Bulgaria	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%		
Croatia	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%		
Czech Rep.	2.2%	2.5%	6.1%	13.9%	23.6%	45.9%	55.3%		
Estonia	0.1%	0.1%	0.2%	0.5%	0.6%	1.2%	2.4%		
Hungary	12.4%	12.5%	13.6%	15.0%	18.4%	24.8%	26.3%		
Latvia	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%		
Lithuania	0.0%	0.0%	0.3%	0.3%	3.9%	5.6%	25.3%		
Norway	6.6%	7.0%	8.5%	10.5%	11.8%	14.6%	16.2%		
Poland	1.1%	1.7%	3.5%	7.4%	11.6%	20.5%	26.3%		
R.Moldova	0.8%	1.3%	1.6%	2.5%	3.1%	3.2%	3.2%		
Romania	0.3%	0.3%	0.3%	0.9%	1.0%	1.1%	1.1%		
Russia	0.0%	0.1%	0.5%	1.0%	1.3%	2.0%	2.8%		
Slovakia	6.8%	7.6%	8.9%	11.3%	12.5%	18.3%	21.2%		
Slovenia	0.0%	0.2%	0.2%	1.0%	2.4%	2.7%	3.0%		
Switzerland	2.2%	2.3%	2.9%	4.2%	5.2%	9.3%	16.2%		
FYRMacedonia	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%		
Ukraine	2.6%	3.5%	3.8%	7.5%	8.2%	11.1%	11.9%		
F.Yugoslavia	0.0%	0.0%	0.0%	0.0%	0.1%	0.1%	0.1%		
Non-EU	0.6%	0.7%	1.3%	2.2%	2.8%	4.2%	5.4%		
TOTAL	0.00/	1 10/	1 70/	0.70/	2 504		7 20/		
TOTAL	0.9%	1.1%	1.7%	2.7%	3.5%	5.5%	1.2%		

5 Discussion and Conclusions

This paper makes a first attempt to propagate uncertainties through the RAINS model calculations from economic activity to the protection of ecosystems. While the methodology is now developed and an example implementation is available, one should be careful when drawing quantitative conclusions from this analysis. Within the given time it was not possible to include the full range of parameters that contribute to uncertainties in the analysis, so that the results presented in this paper are based on an incomplete representation of existing uncertainties. Furthermore, for many of the factors that are included in the analysis it was only possible to derive preliminary quantifications of their uncertainties, and more work is required to obtain improved estimates.

Given these limitations the discussion and conclusions in this paper are restricted to general features of uncertainties. Numerical results could change in future if additional factors are considered.

5.1 General Observations about Uncertainties

As a general observation we found it much more difficult to quantify uncertainties on a solid basis than, e.g., mean values that are used in traditional deterministic analyses. Therefore, we consider the quantification of the **uncertainties** themselves as the **most uncertain** element in the **uncertainty analysis**.

This observation holds despite the methodological approach, which relies solely on the first and second moments of model parameters (i.e., the means, the variances and correlations) and, in contrast to many other approaches to uncertainty analysis, does not require assumptions about distributions, which are even more difficult to establish on a firm basis.

Furthermore, we found it most difficult to quantify (in several cases even the sign of) correlations between input parameters. As a consequence we could only consider a very limited number of correlations, accepting that this limitation could have bearings on the conclusions of the analysis.

The error propagation methodology developed for this paper is only applicable to additive and multiplicative models. It cannot be used for non-linear models, e.g., for determining uncertainties of critical loads from input parameters, or more generally, for any process involving ranking/substituting of options.

It is pointed out in this paper at several places that quantitative uncertainty estimates can only be computed for specific model outputs. General notions like 'the uncertainty of a given model' do not appear particularly useful concepts. Different types of model output have different uncertainties, as demonstrated, e.g., in the case of sectoral and national total emissions. This also reinforces the basic concept that each model has its specific purpose for which it was constructed and for which the control of uncertainties is a critical issue. Using the same model for other endpoints might put the uncertainties in a completely different context and requires careful analysis of these implications.

5.2 Uncertainty of Emission Estimates

It was found that the uncertainties in calculations of emissions, which add up a large number of multiplicative operations for individual sources, are strongly determined by the potential for error compensation. This potential is larger - and therefore the uncertainties are smaller – if more elements of similar sizes are included and if there is no (emission) source that makes a dominating contribution.

Therefore, in general RAINS model estimates of sectoral emissions are more uncertain than national total emissions. The error compensation leads to the situation that in many countries levels of national SO_2 emissions turn out to be more uncertain than those of NO_x and even NH₃, despite uncertainties in many of the input parameters for NO_x and NH₃ calculations being larger than those for SO_2 .

This finding has an implication on the optimal design of emission inventories, suggesting that more resolved emission inventories should be associated with less overall uncertainties. However, the potential for such improvements is limited by the associated need for additional information at the more resolved level with equal quality. Simple disaggregation of sectors without additional genuine information would just introduce strongly correlated terms in the analysis, which in turn will not influence the uncertainties of the overall estimate.

Real improvements in emission inventories are inextricably linked to the availability of additional information and deeper insight into the correlations of parameters.

5.3 Uncertainties of Deposition Estimates

For practical reasons the analysis conducted for this paper could not explore the full range of potential factors that contribute to the uncertainties in the estimates of atmospheric dispersion of pollutants. Further insights could be gained through additional analysis, e.g., with the EMEP dispersion model.

The analysis, which focused on the inter-annual meteorological variability, shows that also for deposition estimates error compensation is an important mechanism with direct impacts on the uncertainties of results. Deposition estimates are more uncertain if deposition at a given site is dominated by the emissions of single source (region), e.g., at the Kola Penninsula and in Romania. Also an uneven distribution of emissions within a country (e.g., if there is only one large power station making a dominant contribution to national emissions) leads within the country to larger uncertainties in the deposition field due to the inter-annual meteorological variability.

In general, the combined uncertainties of emission estimates and of the inter-annual meteorological variability leads to similar uncertainties of sulfur and reduced nitrogen (NH_3) deposition fields, while for oxidized nitrogen uncertainties turn out to be slightly lower.

The analysis reveals an interesting aspect showing lower uncertainty in the field of total nitrogen deposition than for the two individual components. This is caused by a negative correlation between NO_x and NH_3 deposition, which was identified from the EMEP model calculations for areas close to emission sources as a consequence of meteorology and short-term chemical reactions. Sunny and warm weather increases the conversion rate from NO to NO_3^- , which shortens the travel distance of nitrogen oxides and increases local deposition. For ammonia, larger rates of local (wet) deposition occur in wet weather conditions.

5.4 Uncertainties of Estimates on Ecosystems Protection

The analysis presented in this paper is based on Europe-wide extrapolations of parameter uncertainties of German data, so that the conclusions must be considered as preliminary. Furthermore, the assumption of perfect correlation between critical loads and protection isolines within a grid cell might be changed in the future.

For estimates of ecosystems protection the spread of critical loads within a grid cell appears to have stronger impact on the resulting uncertainties and possible error compensation (as long as perfect correlation is assumed). This means in cases where countries report only few critical loads for grid cells or where these critical loads are in a similar range, estimates of ecosystems protection are rather uncertain since, e.g., a small change in these data or in deposition might change the protection status for many ecosystems. Furthermore, it is important to point out that the traditional deterministic calculations of the RAINS model represent the median of the probability distribution and thereby assume a 50 percent probability of the achievement of the environmental targets. It is clear from the calculations that there is a significant uncertainty interval around the median and, depending on the level of confidence one puts into the calculations, the achievement of the original policy target appears in a different light.

As a conclusion, setting of interim or long-term environmental policy targets should not only address the desired level of protection but at the same time also consider the certainty with which this level should be achieved. As shown above the uncertainty range is considerable, and it needs to be explored how different confidence levels will influence the economic efforts that are needed to attain them.

5.5 Conclusions

A methodology to propagate uncertainties in model parameters through the calculation chain of the RAINS integrated assessment model is now available. Initial uncertainty analyses highlight the potential for error compensation, so that in many cases calculation model results are more certain than some of the input parameters.

Obviously, there is ample scope for reducing the uncertainties of model calculations. In all cases, however, only additional quality controlled information from countries can lead to substantial improvements.

6 References

Alcamo J., ApSimon H. and Builtjes P. eds. (1987) *Interregional Air Pollutant Transport: The Linearity Question.* RR-87-20, International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Alcamo J. and Bartnicki J. (1987) A framework for error analysis of a Long-range transport model with emphasis on parameter uncertainty. RR-87-23, International Institute for Applied Systems Analysis, Laxenburg, Austria.

Alcamo J. and Bartnicki J. (1990) *The Uncertainty of Atmospheric Source-Receptor Relationships in Europe*, Atmospheric Environment (1990) 24A, 2169-2189.

Altman A., Amann M., Klaassen G., Ruszczynski A. and Schöpp W. (1996) *Cost-effective sulphur emission reduction under uncertainty*. European Journal of Operational Research (1996) 90, 395-412.

Amann M. and Lutz M. (2000) *The revision of the air quality legislation in the European Union related to ground-level ozone*. Journal of Hazardous Materials 78 (1-3).

Bak J. and Tybirk K. (1998) *The EU Acidification Strategy: Sensitivity of calculated emission ceilings for nitrogen and sulphur for Denmark*, Environmental Pollution (1998) 102, 625-633.

Barkman A. (1997) *Applying the Critical Loads Concept: Constraints Introduced by Data Uncertainty.* Reports on Ecology and Environmental Engineering, Rep. 1:1997. Lund University, Lund, Sweden.

Barkman A. (1998) *Critical loads – assessment of uncertainty*. Reports in ecology and environmental engineering 1:1998, Lund University, Sweden.

Barret K. and Sandnes H. (1996) *Transboundary Acidifying Air Pollution calculated transport and exchange across Europe, 1985-1995.* In: Barret K., Berge E. (eds.) Transboundary Air Pollution in Europe. MSC-W Status Report 1996, Meteorological Sythesizing Centre - West, Norwegian Meteorological Institute, Oslo, Norway.

Bertok I., Cofala J., Klimont Z., Schöpp W. and Amann M. (1993) *Structure of the RAINS 7.0 Energy and Emissions Database*. WP-93-67, International Institute for Applied Systems Analysis, Laxenburg, Austria.

Cofala J. and Syri S. (1998a) Sulfur Emissions, Abatement Technologies and Related Costs for Europe in the RAINS Model Database. IR-98-035 International Institute for Applied Systems Analysis, IIASA, Laxenburg, Austria.

Cofala J. and Syri S. (1998b) Nitrogen Oxides Emissions, Abatement Technologies and Related Costs for Europe in the RAINS Model Database. IR-98-088 International Institute for Applied Systems Analysis, IIASA, Laxenburg, Austria.

Duerinck J. (2000) Statistical properties of national emission reduction cost functions. Pollution Atmospherique 10/2000.

Hettelingh J.-P. (1989) Uncertainty in Modeling Regional Environmental Systems: The generalization of a watershed acidification model for predicting broad scale effects, Free University of Amsterdam, Netherlands.

Hettelingh J.-P. (1990) Uncertainty in Modeling Regional Environmental Systems. The Generalization of a Watershed Acidification Model for Predicting Broad Scale Effects. RR-90-3, International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Isherwood, K.F. (2001) *Urea: codes of good practice*, in UN/ECE Ammonia Expert Group Meeting, Proceedings of the Meeting from 18-20 September 2000. Environmental Documentation No. 133. Swiss Agency for the Environment, Forests and Landscape (SAEFL), Berne, 2001.

Klimont Z., Amann M. and Cofala J. (1998) *Estimating Costs for Controlling Emissions of Volatile Organic Compounds (VOC) from Stationary Sources in Europe*. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Milton J. and Arnold J. (1995) Introduction to probability and statistics. Princibles and Applications for Engineering and the Computing Sciences, Third edition. McGraw-Hill, Singapore.

Posch M. et al. (2001) Calculation and mapping of critical thresholds in Europe. Status Report 2001, Coordination Center for Effects, RIVM, Netherlands.

Posch M., Hettelingh J.-P., de Smet P.A.M. and Downing R.J. (eds.) (1999) *Calculation and mapping of critical thresholds in Europe*. Status Report 1999, Coordination Center for Effects, National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

SAEFL (2001) UN/ECE Ammonia Expert Group Meeting, *Proceedings of the Meeting from* 18-20 September 2000. Environmental Documentation No. 133. Swiss Agency for the Environment, Forests and Landscape (SAEFL), Berne, 2001.

Schönfeld P. (1971) Methoden der Ökonometrie, Band II, Stochastische Regressoren und simultane Gleichungen. Verlag Franz Vahlen, München.

Schöpp W., Amann M., Cofala J., Heyes C. and Klimont Z. (1999) *Integrated Assessment of European Air Pollution Emission Control Strategies*. Environmental Modeling and Software 14(1).

Schöpp W., Heyes C. and Amann M. (2001) *The Treatment of Uncertainties in RAINS Model Calculations Leading to the EU Directive on National Emission Ceilings and the UN/ECE Acidification/Eutrophication/Ozone Protocol*. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Simpson D. (1992) Long Period Modelling of Photochemical Oxidants in Europe : A) Hydrocarbon Reactivity and Ozone Formation in Europe. B) On the Linearity of Country-to-Country Ozone Calculations in Europe. EMEP MSC-W Note 1/92, Norwegian Meteorological Institute, Oslo, Norway.

Simpson D. (1993) Photochemical Model Calculations over Europe for two Extended Summer Periods : 1985 and 1989. Model Calculations and Comparison with Observations. Atmos. Environ., 27A, No. 6, pp. 921-943.

Sorensen L. (1994a) *Evaluation of Integrated Environmental Models. A Case Study.* Riso National Laboratory, Roskilde, Denmark.

Sorensen L. (1994b) *Environmental Planning and Uncertainty*. Riso National Laboratory, Roskilde, Denmark.

Sutton, M. (1995) *Cultures with Fertilizers – 100100*, in Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, EEA, Copenhagen, 1996.

Syri S., Suutari R. and Posch M. (2000) From emissions in Europe to critical load exceedances in Finland - uncertainty analysis of acidification integrated assessment Environmental Science & Policy (2000) 3, 263-276.

UN/ECE (1999) Control Options/Techniques for Preventing and Abating Emissions of Reduced Nitrogen Compounds. EB.AIR/WG.5/1999/8.

Van der Hoek K. (1995) *Manure Management – 100500*, in Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, EEA, Copenhagen, 1996.

Van Sluijs J. (1996) Integrated Assessment Models and the Management of Uncertainties. WP-96-119, International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Webb J. (2001) A Summary of Field-Scale Tests of applying Slurry by Band Spreading, *Trailing Shoe or Shallow Injection to Reduce Ammonia Emissions*, in UN/ECE Ammonia Expert Group Meeting, Proceedings of the Meeting from 18-20 September 2000. Environmental Documentation No. 133. Swiss Agency for the Environment, Forests and Landscape (SAEFL), Berne, 2001.

7 Annex: Relevant Parts from Statistical Textbooks

All the statistical calculations are based on expected values. For example the mean of the random variables is the expected value of the variable, variance is the expected squared difference between a random variable and its expected value. If we have a random variable χ that has a continuous probability distribution $f(\chi)$ the expected value of χ is

$$E[\chi] = \int_{-\infty}^{\infty} \chi f(\chi) \, \mathrm{d}\chi \tag{1}$$

The following rules of expectations can be proved with the equation (1). Let χ and ψ be random variables and c be any number, then:

- 1. **E**[*c*] = *c*
- 2. $\mathbf{E}[c\chi] = c\mathbf{E}[\chi]$
- 3. $\mathbf{E}[\chi + \psi] = \mathbf{E}[\chi] + \mathbf{E}[\psi]$
- 4. $\mathbf{E}[\chi \psi] = \mathbf{E}[\chi] \mathbf{E}[\psi]$,

if χ and ψ are independent

(2)

The following rules of variation can be proved with the above rules (2). Let χ and ψ be random variables and c be any number, then:

- 1. **Var**[c] = 0
- 2. $\operatorname{Var}[c\chi] = c^2 \operatorname{Var}[\chi]$
- 3. $\operatorname{Var}[\chi + \psi] = \operatorname{Var}[\chi] + \operatorname{Var}[\psi] + 2\operatorname{Cov}[\chi, \psi]$
- 4. $\operatorname{Var}[\chi \psi] = \operatorname{Var}[\chi] \mathbf{E}[\psi]^2 + \operatorname{Var}[\psi] \mathbf{E}[\chi]^2 + \operatorname{Var}[\chi] \operatorname{Var}[\psi]$, if χ and ψ are independent

The method for estimating the uncertainty of the integrated acidification model is based on the definitions of variance, covariance and Pearson coefficient of correlation:

1. Definition of variance:

$$\mathbf{Var}[\boldsymbol{\chi}] = \mathbf{E}[(\boldsymbol{\chi} - \mathbf{E}[\boldsymbol{\chi}])^2] = \mathbf{E}[\boldsymbol{\chi}^2] - \mathbf{E}[\boldsymbol{\chi}]^2 \equiv \sigma_{\boldsymbol{\chi}}^2$$

2. Definition of covariance:

$$\mathbf{Cov}[\boldsymbol{\chi}, \boldsymbol{\psi}] = \mathbf{E}[(\boldsymbol{\chi} - \mathbf{E}[\boldsymbol{\chi}])(\boldsymbol{\psi} - \mathbf{E}[\boldsymbol{\psi}])] = \mathbf{E}[\boldsymbol{\chi}\boldsymbol{\psi}] - \mathbf{E}[\boldsymbol{\chi}]\mathbf{E}[\boldsymbol{\psi}]$$
(3)

3. Definition of coefficient of correlation:

$$\rho_{\chi\psi} = \frac{\operatorname{Cov}[\chi, \psi]}{\sqrt{\operatorname{Var}[\chi]\operatorname{Var}[\psi]}} = \frac{\operatorname{Cov}[\chi, \psi]}{\sigma_{\chi}\sigma_{\psi}}$$

We can notice that the covariance of variable itself is the variance of the variable

$$\mathbf{Cov}[\boldsymbol{\chi}, \boldsymbol{\chi}] = \mathbf{E}[\boldsymbol{\chi}^2] - \mathbf{E}[\boldsymbol{\chi}]^2 = \mathbf{Var}[\boldsymbol{\chi}]$$
(4)

For the uncertainty calculations we estimate the standard deviations (square root of the variation) and the correlation coefficients of the model parameters. In the uncertainty calculations we present the uncertainty as a factor of the model parameter. If the model parameter is assumed to be unbiased the uncertainty factor has expected value one.

To demonstrate how the main equations in the following methods have been derived we study simple sums

$$\sum_{i} a_i \chi_i b_i \psi_i$$
 and $\sum_{p} a_p \chi_p b_p \psi_p$

where *a* and *b* are constants and χ and ψ are random variables (uncertainty factors for a and b) with expected value one. It is necessary to assume that χ and ψ random variables are independent. χ_i and χ_p can be correlated as well as ψ_i and ψ_p for any *i* and *p*. Then the expected value of the sum is

$$\mathbf{E}[\sum_{i} a_{i} \boldsymbol{\chi}_{i} b_{i} \boldsymbol{\psi}_{i}] = \sum_{i} a_{i} b_{i} \mathbf{E}[\boldsymbol{\chi}_{i}] \mathbf{E}[\boldsymbol{\psi}_{i}] = \sum_{i} a_{i} b_{i}$$
(5)

The covariance between two such sums is

$$\operatorname{Cov}\left[\sum_{i}a_{i}\chi_{i}b_{i}\psi_{i},\sum_{p}a_{p}\chi_{p}b_{p}\psi_{p}\right]$$

$$=\operatorname{E}\left[\sum_{i}a_{i}\chi_{i}b_{i}\psi_{i}\sum_{p}a_{p}\chi_{p}b_{p}\psi_{p}\right]-\operatorname{E}\left[\sum_{i}a_{i}\chi_{i}b_{i}\psi_{i}\right]\operatorname{E}\left[\sum_{p}a_{p}\chi_{p}b_{p}\psi_{p}\right]$$

$$=\operatorname{E}\left[\sum_{i}\sum_{p}a_{i}\chi_{i}b_{i}\psi_{i}a_{p}\chi_{p}b_{p}\psi_{p}\right]-\sum_{i}a_{i}b_{i}\operatorname{E}\left[\chi_{i}\right]\operatorname{E}\left[\psi_{i}\right]\sum_{p}a_{p}b_{p}\operatorname{E}\left[\chi_{p}\right]\operatorname{E}\left[\psi_{p}\right]$$

$$=\sum_{i}\sum_{p}a_{i}a_{p}b_{i}b_{p}\operatorname{E}\left[\chi_{i}\chi_{p}\right]\operatorname{E}\left[\psi_{i}\psi_{p}\right]-\sum_{i}\sum_{p}a_{i}a_{p}b_{i}b_{p}$$
(6)

To solve the covariance we have defined the expected value of a product of two random variables. For that we use the definitions of covariance and coefficient of correlation (definitions 3). From the definitions we derive the equation

$$\sigma_{\chi_i} \sigma_{\chi_p} \rho_{\chi_i \chi_p} = \mathbf{E}[\chi_i \chi_{pi}] - \mathbf{E}[\chi_i] \mathbf{E}\chi_p] = \mathbf{E}[\chi_i \chi_{pi}] - 1$$

$$\Leftrightarrow \mathbf{E}[\chi_i \chi_{pi}] = \sigma_{\chi_i} \sigma_{\chi_p} \rho_{\chi_i \chi_p} + 1$$
(7)