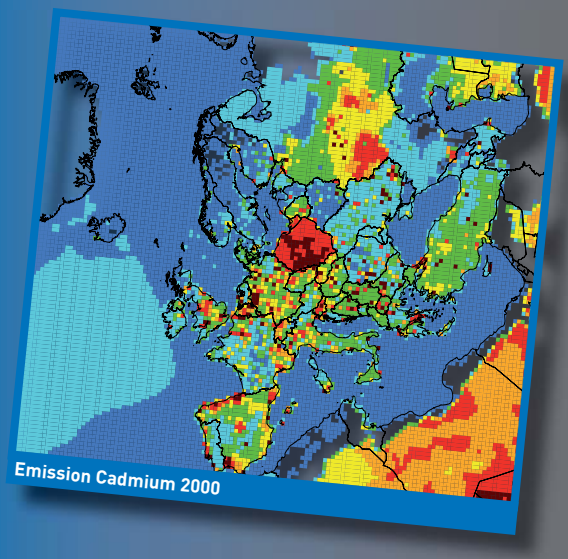
# Heavy Metal Emissions, Depositions, Critical Loads and Exceedances in Europe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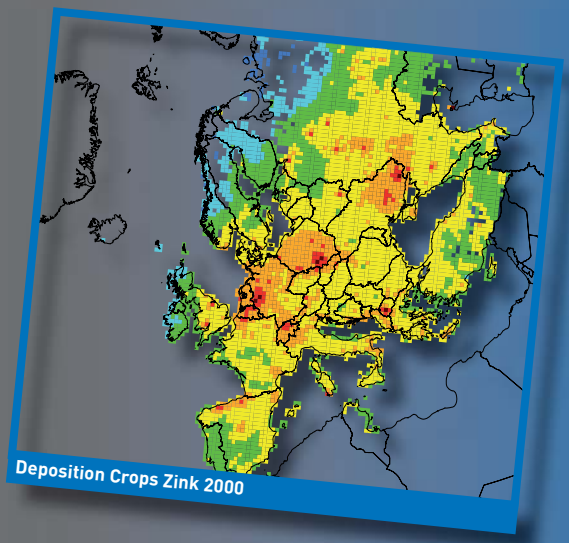
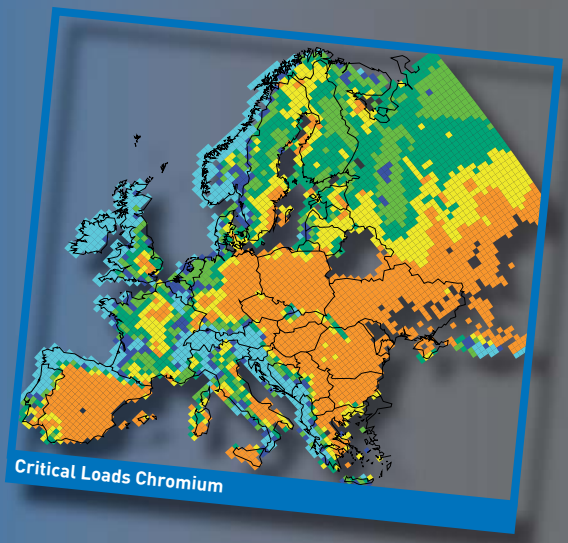
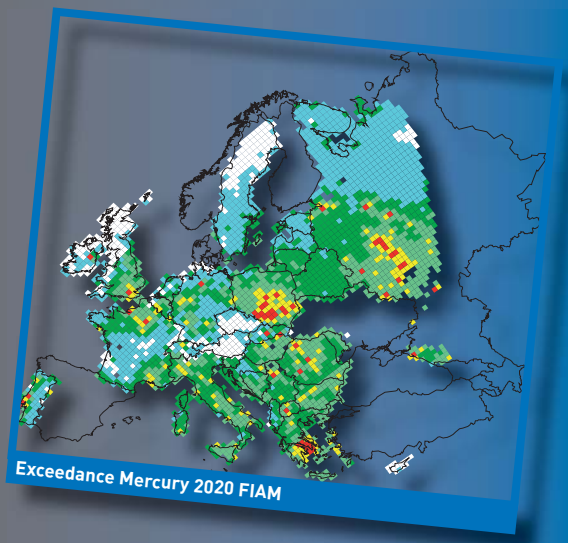

Deposition Crops Zink 2000


Critical Loads Chromium


Exceedance Mercury 2020 FIAM

LRTAP
Long-range Transboundary Air Pollution

TNO Built Environment and Geosciences

MSC-E
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Netherlands Environmental Assessment Agency

wge
Working Group on Effects of the Convention on Long-range Transboundary Air Pollution

VROM

# Heavy Metal Emissions, Depositions, Critical Loads and Exceedances in Europe

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6 VROM

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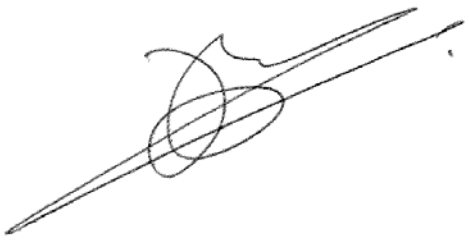
# Preface

The 1998 Heavy Metal (HM) Protocol under the Convention on Long-range Transboundary Air Pollution entered into force 29 December of 2003 implying the start of the review of the Protocol. The focus of the review is on the sufficiency and effectiveness of the Protocol. The Task Force on Heavy Metals was asked by the Executive Body in December 2005 to finalize its 'Sufficiency and Effectiveness Review' (E&S Review) in 2006 in time for the Executive Body meeting in 2006.

An important aspect of the review of the Protocol is to give information of the effects of heavy metals concentrations and depositions on human health and the environment. Does current emissions to air result in effects? The E&S Review covers that on the basis of measurements and exceedance calculations for current emission inventories for the three priority metals, Cadmium, Lead and Mercury. Since TNO has produced emission scenarios for the year 2020, it was tempting to calculate exceedances for heavy metals for these emission scenarios. How would the risks for human health and environment develop when the HM Protocol would be fully implemented by all countries in Europe and what if additional measures on top of the HM Protocol would be taken? For reasons of completeness, the Dutch ministry of environment took the initiative to include chromium, nickel, copper, zinc, arsenic and selenium in the assessment of emissions, depositions and critical load exceedances.

Scenario analysis has been applied in this report to enable a comparison of emission reduction alternatives in terms of risks to human health and the environment, as exceedances of critical loads, for the three priority metals, cadmium, lead and mercury and the six other metals. The results show that even after full implementation of the HM Protocol and additional measures still human health and the environment are at risk. These conclusions are in line with those of the E&S Review and those of the Task Force on Health showing a need for a further reduction of heavy metals.

This work is the result of an excellent collaboration between TNO, EMEP/MSC-E, Alterra, and the ICP-M&M/Coordination Centre for Effects at the Netherlands Environmental Assessment Agency. It is my pleasure to draw your attention to this report and I hope that you will find it useful for the review and possible revision of the Heavy Metal Protocol.



Hans Bolscher
Director Climate Change and Industry
Dutch Ministry of Housing, Spatial Planning and the Environment.

# Summary

This report summarizes knowledge of - and comparison between - heavy metal emission reduction scenario's and resulting depositions and critical load exceedances.

The focus of the report is on the relative assessment of the effects of the three "priority" heavy metals cadmium, mercury and lead, and - more tentatively - of other heavy metals on human health and the environment. Scenario analysis has been applied and described in this report to enable a tentative comparison of emission reduction alternatives in terms of risks to human health and the environment.

The structure of the report follows the cause-effect chain, i.e. the emissions, dispersion, critical loads and critical load exceedances of cadmium, mercury, lead, chromium, nickel, copper, zinc, arsenic and selenium are considered respectively. Three scenarios have been defined to review the risks of impacts to human health and the environment in 2020 in comparison to 2000. For 2020 the scenarios are Current LEgislation (CLE), Full Implementation of the protocol (FI) and Full Implementation plus Additional Measures (FIAM).

The risk of cadmium, mercury and lead, are confirmed in the scenario analysis presented here. The European ecosystem area at risk in 2000 of adjusted national emissions of cadmium, mercury and lead is about 0.34 %, 77 % and 42 % respectively. In 2020 - after full implementation of the Protocol plus additional measures - these areas are reduced to 0.02 %, 74 % and 19 % respectively. The distribution and magnitude of the deposition of mercury and lead in particular puts large areas of European ecosystems at risk of adverse effects both in 2000 and 2020.

The effect based scenario analysis of emissions of chromium, nickel, copper, zinc, arsenic and selenium indicates that a relatively small ecosystem area in Europe is subject to risk of adverse effects of these metals in 2000. For 2020, the application of the CLE, FI or FIAM scenarios leads to the identification of a relative – yet not widely spread – risk of copper, zinc and selenium to remain. The atmospheric depositions of these three metals are computed to cause risk of adverse effects of about 1% or less (in the case of FIAM) of the European ecosystem area in 2020. However, it is noted by EMEP-MSCE that modelled concentrations and depositions of copper, zinc and selenium are significantly underestimated and not recommended for use in exceedance calculations. The tentative use of higher emission data by EMEP-MSCE (see ESPREME project) demonstrated that an improved agreement between modelled and measured depositions in 2000 can be obtained.

Acknowledging that even a small exceedance may result in a future effect on a human or environmental endpoint, the tentative result of the assessment of exceedances of other than priority metals may be considered in line with the conclusions formulated in the Sufficiency and Effectiveness report by the Task Force on Heavy Metals. In the Sufficiency and Effectiveness report it is stated that "Presently, available information indicates that none of these other metals achieve high enough concentrations as a result of long-range atmospheric transport and deposition to cause adverse effects on wildlife and human health".
This report provides further substantiation that the policy focus on priority metals is justifed.

# Table of Contents

# 1. Introduction

The 1998 Heavy Metals Protocol entered into force 29 December 2003 implying the start of the review of the Protocol. On request of the Executive Body in December 2005 the Task Force on Heavy Metals completed its "Sufficiency and Effectiveness" report, in which the current up-to-date knowledge is summarised including data on possible effects caused by heavy metal depositions.

This report provides novel information on emission scenarios, dispersion, critical loads and their exceedances of chromium (Cr), nickel (Ni), cupper (Cu), zinc (Zn), arsenic (As) and selenium (Se). Similar information for cadmium (Cd), mercury (Hg) and lead (Pb) is summarized including references to existing publications.
The report is intended to provide further support to the review and possible revision of the Heavy Metal Protocol.
The report is the result of close collaboration between TNO (emissions), EMEP-MSC/E (dispersion modelling), Alterra (critical loads of "other" metals) and the ICP-M&M/Coordination Centre for Effects (critical loads Cd, Hg and Pb, exceedances).

Chapter 2 describes the methods and data with respect to the source-effect chain consisting of four elements, i.e. emissions, dispersion, critical loads and exceedances in general.
Chapter 3 summarizes comparative-static results of scenario analysis with focus on the relative change of the area at risk of heavy metal deposition. The results are presented for each metal separately. The base year of the scenario analysis is 2000 while 3 scenarios are distinguished to present forecasts of 2020. The scenarios are "Current Legislation" (CLE), " Full Implementation" (FI), and " Full Implementation plus Additional Measures" (FIAM). The risk of impacts of each of the scenario's is geographically illustrated in maps which display the result of comparisons between depositions and critical loads in grid cells of the EMEP domain under the Convention on Long-range Transboundary Air Pollution. In addition country specific areas at risk are calculated for each of the scenarios.
Finally, conclusions and recommendations are in chapter 4.

# 2. Methods and data

## 2.1 Emissions of heavy metals in UNECE-Europe

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### 2.1.1 Introduction

The Dutch Ministry of Housing, Spatial Planning and the Environment (VROM) has commissioned a study to the effectiveness of the UNECE Heavy Metals Protocol and an assessment of possible additional measures with their reductions and costs, based on projections of 2000 emission data to the years 2010, 2015 and 2020. This chapter is based on the results of this study published by Denier van der Gon et al. (2005) and (Visschedijk et al, 2006). The first phase of the study (Denier van der Gon et al., 2005) comprises the construction of an emission inventory for the year 2000, including actualisation of emission data and projections for 2010, 2015 and 2020, geographical allocation of these emissions, efficiency of the current protocols and a preliminary inventory of possible additional reduction measures. Phase II (Visschedijk et al, 2006) comprises an estimation of the emission reduction as well as costs of options for revision of the HM Protocol. In the following sections the assumptions, methodology and results of the studies are briefly presented, a more detailed description and discussion can be found in the respective reports.

### 2.1.2 Scope and limitations of the emission inventories

An emission inventory for Heavy Metals (HM) is made for the year 2000 based on submissions of emission data from the Parties to the Convention on LRTAP. The inventory covers the UNECE territory except Canada and the United States. For the countries, sources or compounds lacking in official submissions, default emission estimates have been prepared and applied to complete the inventory. In the majority of the member countries the relevant experts have been contacted and information on the default emission inventory methodology has been transferred to them and included a feed back by the country experts regarding corrections of official emission data as retrieved by TNO. It is essential to have all relevant source categories covered for all countries to have comparable emission data. Therefore, only official data which have a split at the sector level (e.g. NFR level 1 or SNAP level 1) are used in the compilation because otherwise no indication of completeness of the inventory can be obtained.
Since the study aims to address the effectiveness of the protocol and the potential for additional measures, two important choices are made beforehand; 1st official emission data "overrule" expert estimates and, 2nd re-emissions and illegal emissions are not included in the inventory. The result of these choices may be an underestimation of "real" emissions.

### 2.1.3 Methodology

The HM emission inventory is based on submissions of emission data from the Parties to the Convention on LRTAP as available at http://webdab.emep.int as of March 2005. However, country reportings are not complete. So, next to this official emissions database, a default or "TNO reference" database is made to fill gaps for species and/or sources where country submissions lacked data. The methodologies followed to obtain the official emissions database and the reference database are described in detail by Denier van der Gon et al. 2005)

and summarized below. To create a final dataset the official emission values are merged with the reference emission inventory to get complete coverage of all countries and all sectors in the UNECE-Europe region. Official emission data are only discarded and replaced by TNO reference data in rare cases when: 1) the difference between official and expert estimates could not be understood and, 2) the source in question contributed significantly to the total emissions of the particular substance (> 10%) and, 3) the choice for official emission data would alter the regional emission pattern and outcome of the key source analysis

### 2.1.4 Source categories

The extensive collection of source types in both the Atmospheric Emission Inventory Guidebook (EMEP/CORINAIR 2003) and the HM Protocol Technical Annexes is the basis of the source selection for the inventory. However, for a pragmatic implementation of the inventory and presentation of the results the individual sources distinguished in the inventory are aggregated on a higher level by source category (Table 2.1.1). A detailed breakdown of the country emissions by source and fuel categories is available on CD-ROM in Denier van der Gon et al. (2005).

**Table 2.1.1** Source categories defined in the HM inventory

| Sector Code | Description |
|---|---|
| PHP | Public heat and power; Excludes refineries |
| RCO | Residential, commercial and other combustion; Includes combustion in agriculture |
| IND | Industry; Includes both combustion and process emission, and refineries and fossil fuel production |
| SPU | Solvent and product use; New and existing stocks; Includes wood preservation |
| ROT | Road transport |
| NRT | Non-Road transport |
| WAS | Waste disposal |
| AGR | Agriculture; Excludes combustion emission in agriculture |
| TOTAL | Total of all sectors |

### 2.1.5 Emission factors for 2000

The emission factors are obtained from the latest EMEP CORINAIR guidebook (EMEP/ CORINAIR, 2003) and the European Emission Inventory for HM and POP for 1990 (Berdowski et al., 1997). If no emission factors are available other general guidebooks are scanned e.g., PARCOM ATMOS emission factor manual (Van der Most and Veldt, 1992). Updated values are used if available and appropriate. However, no additional research is undertaken to revise emission factors.

### 2.1.6 Emission projection to future years

Projections of emissions from 2000 to 2010, 2015 and 2020 require assumptions on the penetration of new technologies and on fuel quantities. The penetration of new technologies and better fuel qualities into the system is not explicitly included in the reference database and not available for the official database. To overcome this problem scaling factors are used and indexes are developed for the future years that can be applied to the year 2000 inventory. All emissions within a specific source sector (Table 2.1.1) are scaled to activities and fuels from the reference inventory. The projection of activity data is, as much as possible, based on the

baseline scenarios developed in the framework of the Clean Air for Europe (CAFE) program (Amann et al., 2005). The year 2000 emissions have been projected to the years 2010, 2015, and 2020 following two policy scenarios:

1. Base Line scenario with Current LEgislation and Current Ratification of the UNECE HM Protocol (CLE)
2. Base Line scenario with Current Legislation and Full Implementation of the UNECE HM Protocol. (FI)

## 2.1.7 Emission maps

For the distribution of national country emission totals over the EMEP 50 x 50 km2 grid, both point source information, population density and land use data have been used. Point source information has been applied for the distribution of emissions from the energy transformation sector (including refineries), the industrial combustion (only larger plants ~ > 50 MWth), the iron and steel industry, the non-ferrous metal industry, the cement industry (for central and eastern Europe), petrochemical industry and municipal waste incineration (for Western Europe. All other categories are distributed using population density distribution.

## 2.1.8 Results and discussion

### *2.1.8.1 Heavy metal emissions in 2000 and indicative comparison to 1990*

The result of the European emission inventory for HM in 2000 aggregated to national total emissions is presented in Table 2.1.2. The origin of the national emission data in Table 2.1.2 is indicated by the formatting. In some cases the national official emission reported in Table 2.1.2 (bold figures) is a slightly modified version of such an official figure because one or more minor source categories were not reported or erroneously reported and have been completed by using a TNO expert estimate to achieve completeness. This is marked as “based on official data” in Table 2.1.2 because the modification results only in minor changes from the official figure and the emission estimate is still based on the official submission. In fact such an added TNO expert may even be a zero emission for a particular source category.
For a pragmatic implementation of the inventory and presentation of the results the individual sources distinguished in the inventory are aggregated on a higher level by source category (Table 2.1.1). A detailed breakdown of the country emissions by source and fuel categories is available in Denier van der Gon et al. (2005)

An indicative comparison between the year 2000 HM emissions and the previous TNO 1990 HM inventory (Berdowski et al, 1990) showed that between 1990 and 2000 the emission of Cd, Hg and Pb decreased with ~40%, ~30% and ~ 65%, respectively (Table 2.1.3, Table 2.1.4. ). Please note: the total emissions reported in Table 2.1.3 and Table 2.1.4. do not match exactly because the total UNECE-Europe domain covered by Table 2.1.4 includes more countries because the Berdowski et al. (1997) emission estimates for 1990 do not cover all the countries that are presently in UNECE-Europe. In Table 2.1.4 countries not covered by Berdowski et al. (1997) are represented by their year 2000 emissions, this makes the total emissions higher than the sum of emissions reported in Table 2.1.3 Hence, also emission reduction in the year 2000 relative to 1990 is slightly different.
The HM Protocol focuses on three priority metals (Cd, Hg and Pb) but as a result of the emission reduction measures for the priority HM that the emissions of As, Cu, Ni and Zn are simultaneously reduced with 57%, 53%, 65% and 29%, respectively (Table 2.1.4 ). For Se no 1990 emission data were available and no relative emission reduction could be calculated. chromium emissions in 2000 are estimated to be at the same level as 1990. Considerable Cr emission reductions in many countries are counterbalanced by increasing emissions in others;

chromium emissions are only expected to significantly decrease if all countries implement the HM Protocol.

By contrast the decade 2000-2010 (assuming current ratification of the HM Protocol and compared to the period 1990-2000) is expected to only bring about a large emission reduction for Pb due to the phase out of leaded petrol. However, full implementation of the HM protocol by all UNECE-Europe countries would bring about considerable HM emission reductions.

***Table 2.1.2*** *Heavy Metal emisisons in the year 2000 (emissions based on official data in bold)*

| ISO3 | Cd | Hg | Pb | As (kg/yr) | Cr | Cu | Ni | Se | Zn |
|---|---|---|---|---|---|---|---|---|---|
| ALB | 199 | 203 | 43183 | 175 | 560 | 1176 | 5835 | 127 | 4144 |
| ARM | 129 | 164 | 8274 | 96 | 371 | 553 | 4127 | 153 | 2004 |
| AUT | **1427** | **1151** | **13853** | 2794 | 11331 | 32595 | 43058 | 1826 | 187706 |
| AZE | 2291 | 984 | 12263 | 2285 | 5697 | 4767 | 102477 | 2043 | 11643 |
| BEL | **2750** | **2260** | **133799** | **3840** | **24950** | **26850** | **52690** | **4320** | **166330** |
| BGR | **10986** | **4186** | **214143** | 3474 | 7045 | 18703 | 26420 | 12384 | 130185 |
| BIH | 1675 | 1982 | 96921 | 2239 | 1749 | 9166 | 7070 | 883 | 68974 |
| BLR | **1394** | **363** | **51039** | **3360** | **6308** | **14472** | **94446** | 5462 | **196768** |
| CHE | **2176** | **2630** | **113566** | 798 | 3517 | 21641 | 7388 | 311 | **558285** |
| CYP | 572 | 593 | **73973** | 629 | 1580 | 1614 | 25176 | 521 | 2484 |
| CZE | **2840** | **3840** | **107709** | 11634 | 16633 | 43703 | 47151 | 15785 | 319506 |
| DEU | 21062 | 56014 | 587641 | 34520 | 73574 | 273268 | 247481 | 30299 | 1657857 |
| DNK | **986** | **4925** | **9574** | **974** | **4340** | **11646** | **14246** | **2050** | **65939** |
| ESP | **15521** | **21788** | **932096** | **56072** | **35929** | **149544** | **256487** | **60781** | **789550** |
| EST | **680** | **553** | **40730** | **9668** | **9686** | **3482** | **7865** | **996** | **52963** |
| FIN | **1400** | **500** | **37500** | **4500** | **28000** | **18700** | **33200** | 5491 | **70600** |
| FRA | **10453** | **13375** | **234097** | **25264** | **259392** | **177165** | **221679** | **14262** | **1441439** |
| GBR | **7249** | **8793** | **192840** | **38022** | **69354** | **48183** | **125288** | **28849** | **413183** |
| GEO | 210 | 253 | 6874 | 173 | 563 | 2834 | 6346 | 129 | 3720 |
| GRC | 2844 | 6650 | 132437 | 3975 | 34974 | 18176 | 99577 | 1965 | 76027 |
| HRV | **1019** | **410** | **146907** | **1068** | **4312** | **9788** | **26550** | **633** | **61059** |
| HUN | **2748** | **4197** | **38659** | **5717** | **6661** | **18731** | **37241** | **1621** | **40231** |
| IRL | 1341 | 1763 | 8754 | 1757 | 3884 | 8776 | 46246 | 1414 | 20476 |
| ISL | 81 | 109 | 197 | 94 | 246 | 428 | 3495 | 51 | 2436 |
| ITA | **11051** | **10156** | **908904** | **42619** | **46207** | **72426** | **107911** | **91478** | **1430866** |
| KAZ | 19239 | 17042 | 600841 | 43818 | 32385 | 218235 | 91478 | 11253 | 1026227 |
| KGZ | 347 | 618 | 61214 | 478 | 854 | 2405 | 3435 | 294 | 17259 |
| LTU | **1367** | **603** | **16121** | **792** | **2405** | **6745** | **26586** | 1653 | **63877** |
| LUX | **51** | **275** | 3368 | **79** | **342** | **1252** | **680** | **24** | **36697** |
| LVA | **589** | **148** | **8230** | **624** | **5731** | **4095** | **11006** | **427** | **56498** |
| MDA | **373** | **146** | **3167** | **477** | **626** | **1573** | **5859** | **1707** | **16515** |
| MKD | **9764** | **1843** | **86962** | 760 | 1279 | 3450 | 9511 | 237 | 439638 |
| NLD | **1158** | **578** | **44070** | **1258** | **5558** | **15537** | **53161** | **2334** | **103445** |
| NOR | **725** | **996** | **6035** | **2457** | **8814** | **19329** | 56660 | 496 | 61777 |
| POL | **50400** | **25600** | **647499** | **50400** | **84300** | **374500** | **251600** | 41845 | **2172999** |
| PRT | 3241 | 6796 | 39461 | 4515 | 12371 | 21227 | 93698 | 24630 | 121127 |
| ROM | 17368 | 9158 | 604363 | 4602 | 11758 | 25813 | 82092 | 9715 | 679514 |
| RUS | **111514** | **80122** | **5861767** | 125475 | 1400582 | 800316 | 1368371 | 74454 | 4831987 |
| SVK | **7248** | **4371** | **74342** | **11219** | **8059** | **23685** | **23572** | **7075** | **59189** |
| SVN | **1542** | **644** | **37459** | 789 | 1387 | 4463 | 4352 | 404 | 25219 |
| SWE | **425** | **746** | **11811** | **594** | **6462** | **15116** | **17715** | **569** | **92328** |
| TUR | 16640 | 18247 | 764867 | 15463 | 40463 | 90054 | 227099 | 20775 | 506920 |
| UKR | 23682 | 22334 | 1703249 | 30334 | 64528 | 198395 | 147555 | 17224 | 1298408 |
| YUG | 8653 | 5484 | 299771 | 5168 | 4824 | 31223 | 19819 | 2372 | 119362 |
| Total tonnes/y | 377 | 344 | 15021 | 555 | 2350 | 2846 | 4144 | 501 | 19503 |

***Table 2.1.3**. Emissions of cadmium, mercury and lead for 1990 (Berdowski et al., 1997) and the year 2000 (Denier van der Gon et al. 2005) and the relative change over the decade 1990-2000.*

| ISO3 | Cd | | | Hg | | | Pb | | |
|---|---|---|---|---|---|---|---|---|---|
| | 1990 | 2000 | Change | 1990 | 2000 | Change | 1990 | 2000 | Change |
| | Tonnes/yr | | (%) | Tonnes/yr | | (%) | | | (%) |
| ALB | 0.6 | 0.2 | -69 | 0.5 | 0.2 | -60 | 33.4 | 43.2 | 29 |
| AUT | 5.1 | 1.4 | -72 | 4.3 | 1.2 | -73 | 215.0 | 13.9 | -94 |
| BEL | 9.9 | 2.7 | -72 | 8.9 | 2.3 | -74 | 716.0 | 133.8 | -81 |
| BGR | 8.4 | 11.0 | 31 | 6.9 | 4.2 | -39 | 317.0 | 214.1 | -32 |
| BIH | 0.4 | 1.7 | 315 | 0.2 | 2.0 | 853 | 8.6 | 96.9 | 1023 |
| BLR | 6.6 | 1.4 | -79 | 0.1 | 0.4 | 299 | 736.0 | 51.0 | -93 |
| CHE | 4.2 | 2.2 | -49 | 6.8 | 2.6 | -61 | 520.0 | 113.6 | -78 |
| CYP | 0.2 | 0.6 | 190 | 0.3 | 0.6 | 94 | 0.9 | 74.0 | 8065 |
| CZE | 12.0 | 2.8 | -76 | 9.3 | 3.8 | -59 | 337.0 | 107.7 | -68 |
| DEU | 31.5 | 21.1 | -33 | 113.0 | 56.0 | -50 | 2347.0 | 587.6 | -75 |
| DNK | 2.1 | 1.0 | -53 | 6.9 | 4.9 | -29 | 179.0 | 9.6 | -95 |
| ESP | 36.7 | 15.5 | -58 | 20.2 | 21.8 | 8 | 4674.0 | 932.1 | -80 |
| EST | 3.9 | 0.7 | -82 | 2.0 | 0.6 | -73 | 171.0 | 40.7 | -76 |
| FIN | 3.7 | 1.4 | -62 | 3.0 | 0.5 | -84 | 215.0 | 37.5 | -83 |
| FRA | 14.8 | 10.5 | -29 | 32.5 | 13.4 | -59 | 4414.0 | 234.1 | -95 |
| GBR | 24.9 | 7.2 | -71 | 25.6 | 8.8 | -66 | 2703.0 | 192.8 | -93 |
| GRC | 4.5 | 2.8 | -36 | 7.1 | 6.7 | -7 | 505.0 | 132.4 | -74 |
| HRV | 3.2 | 1.0 | -69 | 1.1 | 0.4 | -62 | 466.0 | 146.9 | -68 |
| HUN | 4.6 | 2.7 | -40 | 4.2 | 4.2 | 0 | 639.0 | 38.7 | -94 |
| IRL | 1.6 | 1.3 | -16 | 1.6 | 1.8 | 9 | 134.0 | 8.8 | -93 |
| ISL | 0.2 | 0.1 | -51 | 0.0 | 0.1 | 127 | 6.4 | 0.2 | -97 |
| ITA | 59.8 | 11.1 | -82 | 11.8 | 10.2 | -14 | 1642.0 | 908.9 | -45 |
| LTU | 2.8 | 1.4 | -52 | 0.0 | 0.6 | NA[a)] | 246.0 | 16.1 | -93 |
| LUX | 1.1 | 0.1 | -96 | 0.8 | 0.3 | -64 | 73.5 | 3.4 | -95 |
| LVA | 3.2 | 0.6 | -82 | 0.3 | 0.1 | -56 | 218.0 | 8.2 | -96 |
| MDA | 1.8 | 0.4 | -79 | 1.5 | 0.1 | -90 | 168.0 | 3.2 | -98 |
| MKD | 9.1 | 9.8 | 7 | 1.5 | 1.8 | 24 | 210.0 | 87.0 | -59 |
| NLD | 2.2 | 1.2 | -47 | 2.6 | 0.6 | -78 | 266.0 | 44.1 | -83 |
| NOR | 2.4 | 0.7 | -70 | 2.3 | 1.0 | -57 | 226.0 | 6.0 | -97 |
| POL | 91.6 | 50.4 | -45 | 33.3 | 25.6 | -23 | 1372.0 | 647.5 | -53 |
| PRT | 3.0 | 3.2 | 9 | 5.5 | 6.8 | 24 | 631.0 | 39.5 | -94 |
| ROM | 21.6 | 17.4 | -20 | 7.5 | 9.2 | 22 | 585.0 | 604.4 | 3 |
| RUS | 159.0 | 111.5 | -30 | 86.2 | 80.1 | -7 | 10148.0 | 5861.8 | -42 |
| SVK | 9.7 | 7.2 | -25 | 12.4 | 4.4 | -65 | 166.0 | 74.3 | -55 |
| SVN | 1.0 | 1.5 | 50 | 0.9 | 0.6 | -26 | 123.0 | 37.5 | -70 |
| SWE | 2.0 | 0.4 | -79 | 1.5 | 0.7 | -49 | 537.0 | 11.8 | -98 |
| UKR | 54.2 | 23.7 | -56 | 36.0 | 22.3 | -38 | 3878.0 | 1703.2 | -56 |
| YUG | 8.3 | 8.7 | 4 | 3.9 | 5.5 | 42 | 597.0 | 299.8 | -50 |
| ***Total[b)]*** | ***612*** | ***339*** | ***-45*** | ***463*** | ***306*** | ***-34*** | ***40424*** | ***13566*** | ***-66*** |

[a)] NA = Not Available (division by zero)

[b)] Year 2000 totals do not add up to totals for Table 3.1.1 to Table 3.9.1 because the UNECE Europe region as of 1990 in Berdowski et al. (1997) cover less countries.

*2.1.8.2 Projected Heavy Metal emissions for 2010-2020 under different policy scenarios*

The HM emission database for the year 2000 has been projected to the years 2010 and 2020 following two policy scenarios

- CLE (Base Line scenario with Current Legislation and Current Ratification (as of April 2005) of HM Protocol)
- FI (Base Line scenario with Current Legislation and Full Implementation of HM Protocol)

Full implementation means implementation of, and compliance with, the HM protocol in all UNECE-Europe countries irrespective of current ratification status. It is important to note that the projected data are created using indexes for the year 2000 emissions. This is crucial to get a consistent outcome because official emission data are the starting point but exact activity data as used by the countries cannot be traced. Hence, a separate bottom-up approach to obtain 2010-2020 emissions could result in unexplainable "jumps" in emissions.

***Table 2.1.4*** *Emissions of selected heavy metals in UNECE Europe for 2000 and projected emissions for 2010 and 2020 following policy scenarios CLE (Base Line scenario with Current Legislation and Current Ratification (as of April 2005) of HM protocol) and FI (Base Line scenario with Current Legislation and Full Implementation of HM protocol).*

| Year policy scenario | Cd | Hg | Pb | As | Cr | Cu | Ni | Se | Zn |
|---|---|---|---|---|---|---|---|---|---|
| | Tonnes / yr | | | | | | | | |
| 1990 | 650 | 501 | 41879 | 1284 | 2289 | 6078 | 11930 | NA[b)] | 27659 |
| 2000 | 377 | 344 | 15021 | 555 | 2350 | 2846 | 4144 | 501 | 19503 |
| 2010 CLE | 327 | 328 | 7317[c)] | 449 | 2328 | 2642 | 3750 | 317 | 18025 |
| 2020 CLE | 323 | 326 | 7650[c)] | 438 | 2645 | 2772 | 3426 | 325 | 19006 |
| 2020 FI | 217 | 316 | 5761[c)] | 318 | 900 | 2126 | 2622 | 294 | 13766 |
| 2020 FIAM[d)] | 137 | 184 | 2791 | 211 | 421 | 1719 | 964 | 206 | 11058 |

[a)] 1990 data taken from Berdowski et al. (1997a) for indicative comparison. Countries not covered by Berdowski et al. are represented by their year 2000 emissions

[b)] NA = Not Available.

[c)] Projected Pb data differ from the figures published in Denier van der Gon et al. (2005) due to a later implemented correction

[d)] Full Implementation of the 1998 HM Protocol plus implementation of Additional Measures (package I+II; Visschedijk et al., 2006). The FIAM emissions listed here are an update of those that have used for the assessment of dispersion and exceedances. The reduction potential of additional measures had been overestimated by about 13 percent-points on average.

***Table 2.1.5*** *Heavy metal emission reduction in 2020 upon full implementation of the 1998 HM protocol compared to implementation of the HM protocol in ratified countries (as of April 2005) and autonomous measures.*

| Cd | Hg | Pb | As | Cr | Cu | Ni | Se | Zn |
|---|---|---|---|---|---|---|---|---|
| Tonnes/yr | | | | | | | | |
| -106 | -10 | -1889 | -120 | -1745 | -646 | -804 | -31 | -5241 |

The difference between the HM emissions after full implementation or after implementation in current ratified countries + autonomous measures (Table 2.1.4.) is rather large for Pb, Cr, Cu, Ni and Zn (Table 2.1.5.).
The reductions given in Table 2.1.5. are not uniformly distributed over the UNECE_Europe domain but almost entirely located in the group of countries which have currently no HM Protocol ratification, no 2nd S protocol ratification and are not a member of EU-25. This can be explained. A country that has not ratified the HM Protocol but e.g., is a member of EU25

will have to comply with the EC directives (E.g. IPPC) and thus has to implement rather stringent ELVs because of autonomous measures. Within this particular group of countries the emission reduction is due to implementation of ELVs and/or other obligations as outlined in the HM protocol. For example, the additional emission reduction of ~1900 tonnes Pb/ yr (Table 2.1.5.) upon full implementation of the HM protocol is achieved in the sector Public heat & power (60%) and the sector Industry incl. combustion (40%). From a national/ geographic perspective, the Pb emission reduction is dominated by emission reduction in Russia (58%), Ukraine (19%) and Kazakhstan (15%).

After full implementation and compliance with the HM Protocol the following source sectors are expected to be the main HM emissions sources: Combustion of fossil fuels for heat and power production, Cement Production, Iron and Steel industry, road transport, industrial and residential combustion and production of copper and zinc (Table 2.1.6.). However, in some cases the emission estimates may be inaccurate due to a lack of good data e.g., data on the lead content in unleaded fuel and the amount of mercury in current and future municipal waste.

***Table 2.1.6*** *Relative contribution of source sectors to remaining HM emissions upon full implementation of the HM protocol by all UNECE-Europe countries.*

| Source sector | Cd | Hg | Pb | As | Cr | Cu | Ni | Se | Zn |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | (%) | | | | |
| Public power and heat | 18 | 40 | 6 | 22 | 8 | 5 | 40 | 18 | 10 |
| Residential combustion | 15 | 8 | 4 | 11 | 10 | 9 | 10 | 3 | 7 |
| Industrial combustion and processes | 60 | 46 | 42 | 66 | 75 | 36 | 45 | 77 | 65 |
| Road transport | 4 | 0 | 45 | 0 | 3 | 31 | 2 | 1 | 13 |
| Non-road transport | 0 | 0 | 2 | 0 | 0 | 17 | 2 | 1 | 0 |
| Waste incineration | 2 | 5 | 1 | 1 | 3 | 2 | 0 | 0 | 5 |
| Total | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |

### *2.1.8.3 Emission reduction due to a possible revision of the HM protocol*

The emission projections under full implementation of the HM protocol can be used as an approximation of the remaining HM emissions in the future. Based on a key source analysis and some additional considerations, possible options for further HM emission reduction are suggested for the heat and power production sector, cement production, sinter plants, blast furnaces, electric arc furnaces, basic oxygen furnaces, open hearth furnaces, coke ovens, production of copper and zinc (including imperial smelting) and road transport (fuel composition). An estimation of the possible emission reduction as well as costs of options for revision of the HM Protocol has been made by Visschedijk et al. (2006). Source sectors considered for a possible revision of the HM Protocol have been selected according to the following criteria:

1. The contribution to the total emission after full implementation of the HM protocol of one or more of the three priority metals (Cd, Hg and Pb) exceeds 5%
2. The contribution to the total emission of one or more of the other heavy metals exceeds 15%
3. Emissions from domestic / residential sources are not considered for revision of the HM Protocol

4. Sources that are scheduled for re-evaluation by the TFHM will be included (HM emissions from Chlor-alkali industry and Medical Waste Incineration )

The sources selected by Visschedijk et al. (2006) cover 74-86% of the priority HM emissions and 64-94% of the other HM. This indicates the potential for reduction, not the actual reduction that will be achieved. For all HM the contribution of residential sources is ~10% (3-15%) but residential combustion is not considered for a possible revision of the HM protocol by Visschedijk et al. (2006) because of foreseen difficulties in implementation, legislation and enforcement.
The measures considered for a possible revision of the HM protocol are described in detail by Visschedijk et al. (2006) and have been separated in a package I and package II proposal. Package I focuses on dust removal measures e.g., by tightening of the dust ELVs as proposed by e.g., Rentz et al. (2004) and the BAT documents. This package addresses all HM except Hg. Mercury is often poorly mitigated by measures which focus on dust removal and dust emission limit values because much of the Hg is emitted in the gaseous phase. In general package II measures are more expensive but can – if desired – replace the Package I measure for the specific sources addressed. Here we present the combined result of implementing Package II and, for sources not addressed by Package II, Package I (Table 2.1.4, Table 3.1.1 to Table 3.1.9 in chapter 3). The projected remaining emissions in 2020 upon full implementation of the 1998 HM protocol and implementation of additional Package I and II measures by source sector in UNECE-Europe is given in Table 2.1.7.

***Table 2.1.7*** *Projected emissions of selected heavy metals in 2020 by source sectors upon full implementation of the 1998 HM protocol by all UNECE-Europe countries and implementation of possible additional measures (Package I +II) as described by Visschedijk et al. (2006).*

| Source sector | Cd | Hg | Pb | As | Cr | Cu | Ni | Se | Zn |
|---|---|---|---|---|---|---|---|---|---|
| | Tonnes/yr | | | | | | | | |
| Public heat and power; Excludes refineries | 10 | 25 | 204 | 35 | 18 | 49 | 39 | 26 | 943 |
| Residential, commercial and other combustion; Includes combustion in agriculture | 34 | 25 | 252 | 34 | 90 | 186 | 252 | 10 | 1008 |
| Industry; Includes both combustion and process emission, and refineries and fossil fuel production | 79 | 117 | 2036 | 139 | 257 | 428 | 544 | 163 | 6707 |
| Solvent and product use; New and existing stocks | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| Road transport | 9 | 0 | 105 | 0 | 31 | 661 | 57 | 4 | 1757 |
| Non-Road transport | 1 | 0 | 112 | 1 | 1 | 363 | 64 | 2 | 21 |
| Waste disposal | 5 | 16 | 81 | 2 | 23 | 32 | 8 | 1 | 622 |
| Agriculture; Excludes combustion emission in agriculture | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| Total of all sectors | 137 | 184 | 2791 | 211 | 421 | 1719 | 964 | 206 | 11058 |

### *2.1.8.4 Co-benefits of implementation of the HM Protocol*

It is estimated that full implementation of the HM Protocol will - as a side effect –result in particulate matter (PM) emission reduction; ~ 3.7 Mt TSP (total suspended particles), 1.2 Mt PM10 and 0.28 Mt PM2.5. Compared to the total European PM emissions in 1995, this is ~ 25% of total TSP, 16% of total PM10 and 6% of total PM2.5 emission, with the largest reduction achieved in the power generation sector. In this study the choice is made to first implement autonomous measures (e.g. IPPC directive for the EC25) and than quantify the additional PM reduction due to implementation of the HM protocol. If the procedure would be

followed differently (first HM protocol, than (other) autonomous measures) the side effect of the HM Protocol on PM reduction is much larger. The possible additonal measures after full implementation of the HM Protocol proposed by Visschedijk et al. (2006) will also bring about a reduction of PM emissions. However, the impact is considerably less because within the source sectors most relevant for HM, the 1998 HM protocol already greatly reduces PM emissions. Subsequent more and more advanced PM reduction measures generally have to deal with smaller particles which, from a health perspective may be highly relevant but on a mass basis contribute relatively little to total PM emissions.

### 2.1.9 Conclusions

Full implementation of the HM Protocol is an important step in HM emission reduction. If all countries of UNECE-Europe implement the HM protocol the projected emissions are considerably reduced (20-60%) compared to the year 2000 for all HM except Hg.

- For Pb, Se and As considerable reduction (20~40%) is achieved going from the year 2000 to 2010 following the baseline scenario and current ratification of the HM protocol.
- The lead emissions strongly decline going from 2000 to 2010 due to the phase out of leaded gasoline. The remaining limit value of Pb in fuel causes road transport to remain an important source of Pb. However, it is possible that Pb levels in gasoline could be much lower.
- Emission of Cr is the only HM that is expected to grow in emission compared to 2000. This is due to activity increase in (Ni production) countries that have currently not ratified the Protocol.
- The difference in HM emissions under the two policy scenarios is larger than the emission changes over time within a policy scenario (e.g. going from 2010 to 2020).

The last conclusion illustrates that full implementation of the HM Protocol is an important step in HM emission reduction. The relative small importance of the projection years (2010-2015-2020) can be explained by 1) our assumption that measures following implementation of the HM protocol will be in effect before 2010 (in both policy scenarios) and little additional measures are yet defined for the period after 2010.

The emission reduction for Hg due to implementation of the HM protocol lags behind because much of the Hg emissions are in the gaseous phase and is poorly mitigated by the measures proposed in the HM protocol as they often focus on dust removal. This is also the reason why Hg emission reduction over 1990-2000 has been smaller than for the other two priority HM. Hg emission reduction warrants special attention in any further policy making aiming at HM emissions reduction. In the study on possible revision of the HM Protocol a separate package was defined to address Hg emission which brings about major Hg emission reduction.
The possible measures suggested for a revision of the HM protocol by Visschedijk et al. (2006) appear expensive compared to the 1998 HM protocol. Berdowski et al. (1998) estimated that, when autonomous developments (e.g. the 2nd S Protocol) where taken into account, the annual costs of implementation of the first draft of the UNECE HM Protocol were ~440 MECU (1995). The Package 1 revision of the HM Protocol would bring about annual costs more than ten times as much (7.6 billion € (2000)). For example, the implementation of the draft 1998 HM Protocol would result in overall specific costs of 2 – 3 € (1995) / g Cd avoided (Berdowski et al., 1998) whereas the possibly revised Protocol regarded by Visschedijk et al (2006) has an average costs effectiveness of 120 € (2000) / g Cd avoided. If only measures with specific costs below 10 € / g Cd avoided would be selected for a revision of the HM Protocol ~50% of the total reduction potential (about 40 tonnes Cd) could be reached. Although a revision of the HM Protocol appears costly compared to the

1998 HM protocol, the costs are still well below the costs of implementation of the 2nd S Protocol.

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## 2.2 Deposition modelling

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### 2.2.1 Brief description of the model

MSCE-HM is a three-dimensional Eulerian-type chemical transport model driven by off-line meteorological data. It is developed to evaluate atmospheric transport and deposition of such heavy metals as Pb, Cd and Hg. Pilot parameterisations for some other toxic metals and metalloids like Cr, Ni and As are included as well. The model domain covers the EMEP region (Europe, part of Northern Africa and Middle East, the north-eastern Atlantic and part of the Arctic) with a spatial resolution 50×50 km$^2$.

The vertical structure of the model is formulated in the sigma-pressure (σ-p) coordinate system. The model domain consists of 15 irregular σ-layers and has a top at pressure level equal to 100 hPa. The layers are confined by surfaces of constant σ and do not intersect the ground topography. The vertical grid structure of the model domain is shown schematically in Figure 2.2.1. The midlevel of the lowest σ-layer approximately corresponds to around 40 m. The top of the model domain can be roughly estimated at 15 km.

The model takes into account key processes governing behaviour of heavy metals in the atmosphere and their deposition to the ground. These include emissions, advective transport, turbulent mixing, wet and dry removal, mercury chemical transformations both in gaseous and aqueous phases. Schematically these processes are depicted in Figure 2.2.2.

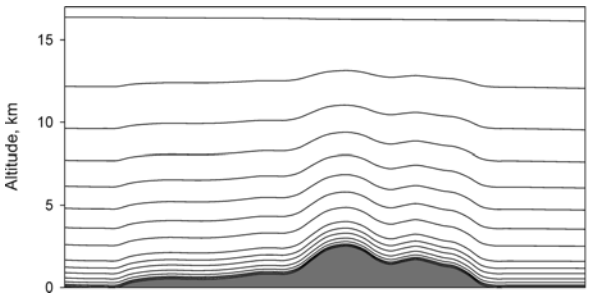


***Figure 2.2.1*** *Vertical grid structure of the model domain. The curves show boundaries of σ-layers*

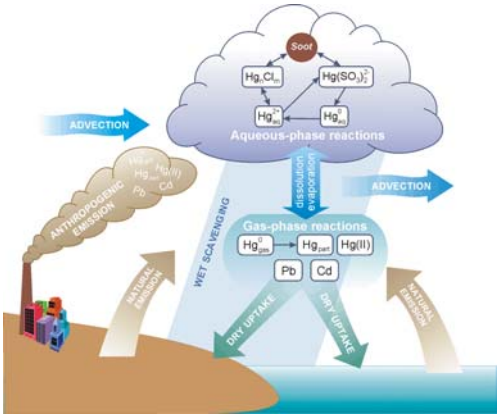


***Figure 2.2.2*** *The model scheme of heavy metal behaviour in the atmosphere*

Advective and vertical transport is evaluated by the Bott scheme (Bott, 1989a; 1989b, 1992). Turbulent mixing is approximated by a second-order implicit numerical scheme. Lead and cadmium are assumed to be transported in the atmosphere only as a part of aerosol particles. Chemical transformations of these metals do not change removal properties of their particles-carriers. Physical and chemical transformations of mercury include dissolution of gaseous elemental Hg in cloud droplets, gas-phase and aqueous-phase oxidation by ozone and chlorine, aqueous-phase formation of chloride complexes, reactions of mercury ion reduction through the decomposition of sulphite complex, and adsorption by soot particles in droplet water. The dry deposition scheme is based on the resistance-analogy approach. Modelled dry deposition velocity depends on surface type (forests, arable lands, water etc.) and atmospheric

conditions (atmospheric stability, wind velocity etc). At present the model is capable of calculating dry deposition fluxes to 18 categories of land cover. The model distinguishes in-cloud and sub-cloud wet scavenging. Boundary concentrations of heavy metals are set along outer boundaries of the EMEP region and updated once a month. Mercury concentrations at the domain boundaries are derived by means of hemispheric-scale modelling. The concentrations of lead and cadmium are based on monitoring data. See Travnikov and Ilyin (2005) for a more technical description of the model.

A special study was undertaken to evaluate uncertainties of the modelling results (concentrations in air, in precipitation, total depositions) caused by uncertainties of model parameters and input data (Travnikov and Ilyin, 2005). The intrinsic uncertainties (uncertainties that exclude emission data uncertainties) and their range over the EMEP domain for lead and mercury are summarized in Table 2.2.1.

**Table 2.2.1** Model intrinsic and the overall uncertainties of the main model output parameters

| Output parameter | Uncertainty, % | Range, % |
|---|---|---|
| *Lead* | | |
| Air concentration | 43 | 22 - 64 |
| Concentration in precipitation | 40 | 20 - 57 |
| Total deposition | 33 | 19 - 49 |
| *Mercury* | | |
| TGM concentration | 19 | 16 - 22 |
| Concentration in precipitation | 53 | 29 - 74 |
| Total deposition | 39 | 20 - 57 |

The reliability of the MSCE-HM model was analysed at the workshop (Moscow, October 2005) of the Task Force of Measurements and Modelling, carried out in the framework of the model review procedure (TFMM Workshop minutes, 2005). The main conclusion of the workshop was that “The MSCE-HM model is suitable for the evaluation of the long-range transboundary transport and depositions of HMs in Europe”.

The workshop also formulated a number of recommendations to improve the model and its input data. One of the recommendations was to develop mechanisms of emissions driven by meteorological processes. Following up on this recommendation, MSC-E has developed a preliminary scheme to calculate heavy metal emissions caused by wind re-suspension from land surfaces and emissions of aerosol-bound metals from sea surfaces.

A detailed description of re-suspension parameterisation is available in Gusev et al. (2006). According to the model description of this process, re-suspension depends mainly on near-surface wind magnitude, soil characteristics and the concentration of heavy metals in soils and water. This section focuses on main results derived from re-suspension parameterisations. Estimates of re-suspension of particle-bound heavy metals from soil and seawater were performed for Europe and adjacent territories in 2000. Spatial distributions of the annual re-suspension flux of Pb, Cd, As, Cr, and Ni are presented in Figure 2.2.3. In general, the re-suspension fluxes from the soil are significantly higher than those from seawater for all the metals. High re-suspension fluxes were obtained from desert areas of Africa and Central Asia because of significant dust production in these regions. Elevated fluxes are also characteristic for some countries of Western, Central, and Southeastern Europe, which are conditioned by a combination of a relatively high concentration in soil and significant dust suspension from urban and agricultural areas. Metals bound to re-suspended particles may have purely natural origins or come from previous long-term (historic) depositions of metals emitted by

anthropogenic sources. At present it is not possible to distinguish between these two components. Therefore, the release of metals to the atmosphere due to re-suspension will also be referred to as natural and historical emission.

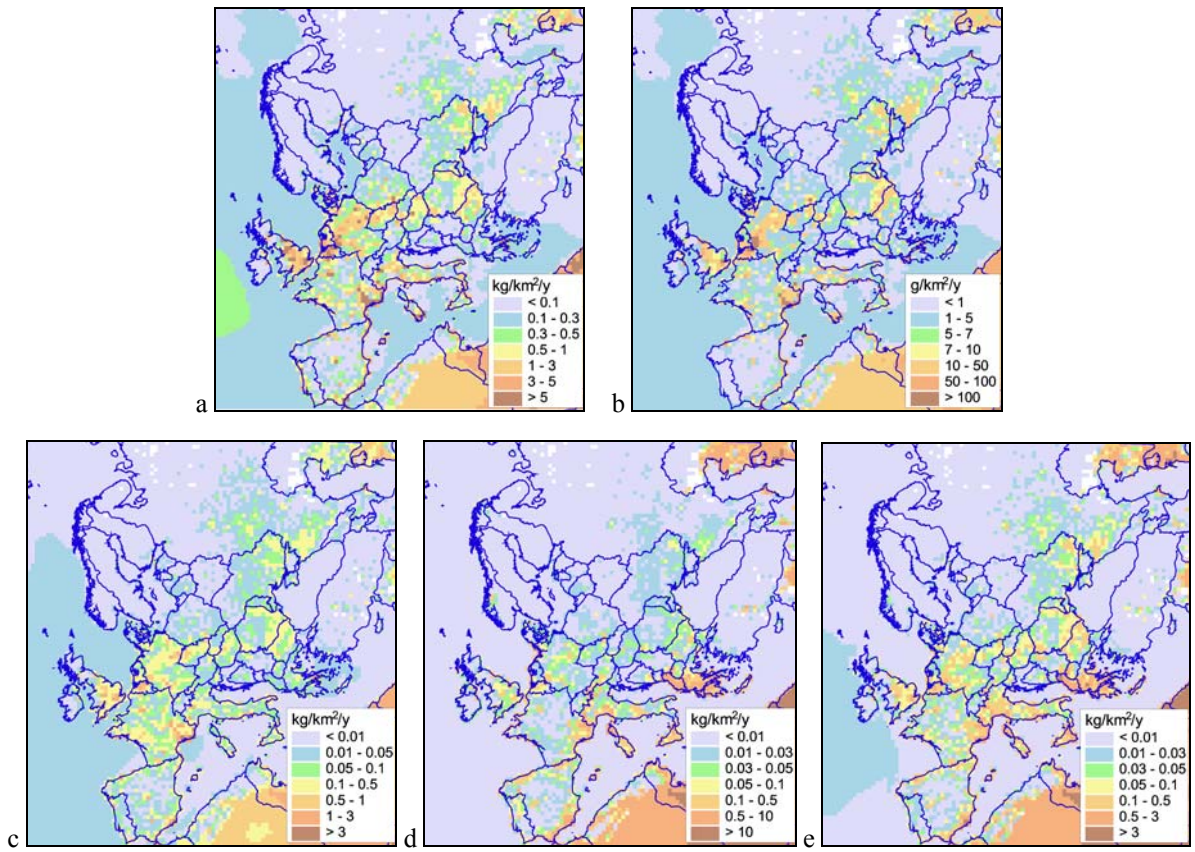


***Figure 2.2.3*** *Spatial distribution of annual resuspension flux of heavy metals in Europe in 2000: (a) – Pb; (b) – Cd; (c) – As; (d) – Cr; (e) – Ni*

Aggregated values of lead re-suspension from soil in different European countries are presented in Figure 2.2.4a along with total anthropogenic emissions based on official data. As can be seen the estimated contribution of the re-suspension of Pb is comparable or even higher than anthropogenic emissions in such countries as Italy, France, Germany, Greece, Spain, the United Kingdom etc., where observed concentration of this metal in soil considerably exceeds its average natural content in the Earth's crust (Figure 2.2.4b) (see Gusev et al, 2006 and references therein). The most probable reason for this is long-term accumulation of historical depositions.

Contrary to lead, cadmium re-suspension from soil insignificantly contributes to the total emission of this metal in most European countries (Figure 2.2.5a). The reason for this is the relatively low cadmium concentrations measured in European soils. Only in a few countries of Europe (France, Italy, Greece, Belgium etc.) mean topsoil concentration noticeably exceeds cadmium natural content in the crust, and natural and historical emissions are comparable with anthropogenic ones (Figure 2.2.5b).

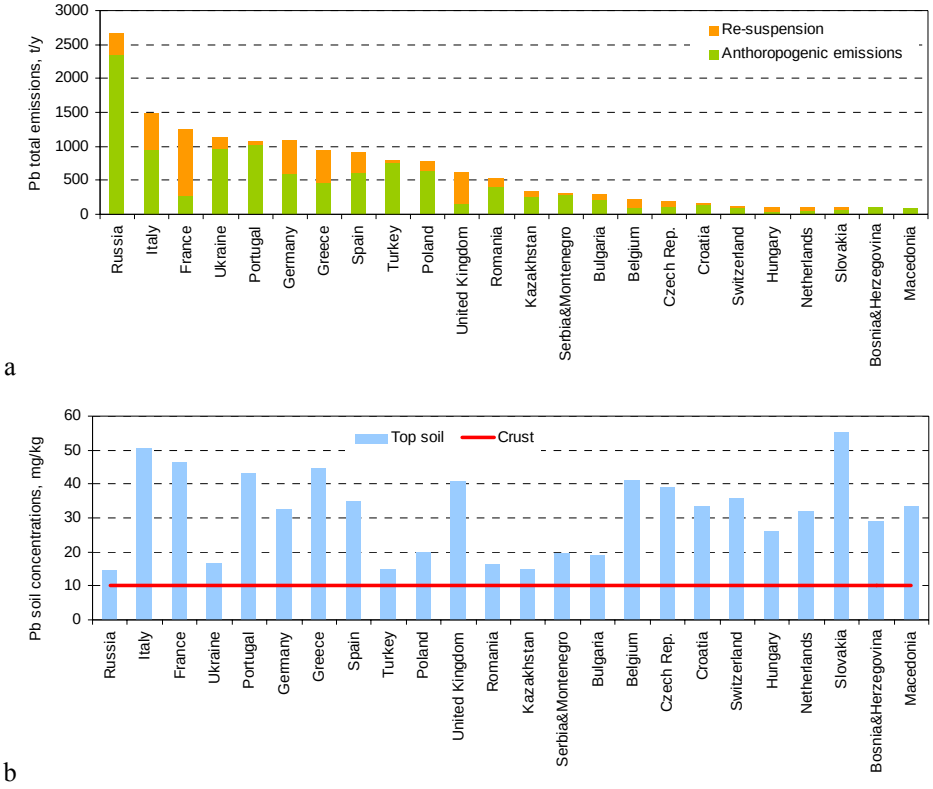


***Figure 2.2.4*** *Lead total anthropogenic emissions and resuspension from soil (a) and average topsoil concentration (b) in some European countries*

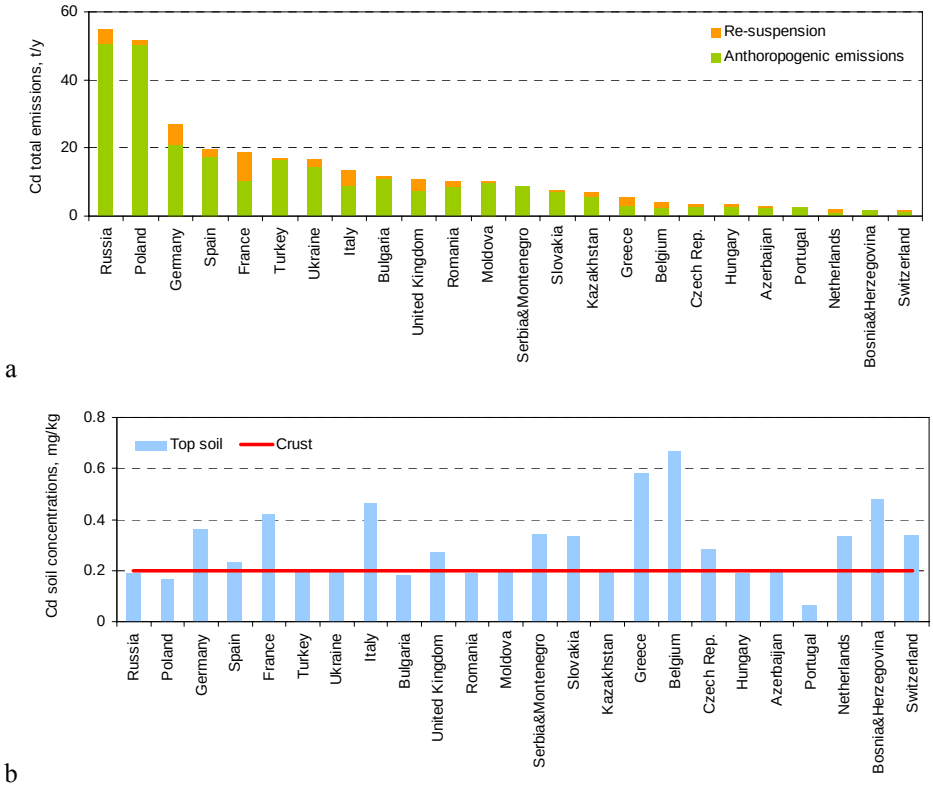


***Figure 2.2.5*** *Cadmium total anthropogenic emissions and resuspension from soil (a) and average topsoil concentration (b) in some European countries*

Pilot parameterization for natural and historic emission of zinc, copper and selenium has also been developed. The approach to quantify these emissions is similar to that used for Pb, Cd, As, Ni and Cr. The exception is that instead of spatially distributed soil concentrations of metals uniform values were used. The currently used parameterization of natural and historic emissions of heavy metals could be considered as preliminary. MSC-E is planning to continue its activity on updating the parameterization of these emissions. In the future, the parameterization can be revised and improved.

### 2.2.2 Heavy metal emission scenarios

In order to calculate depositions to various ecosystems in Europe, four emission scenarios of Pb, Cd, Hg, As, Ni, Cr, Cu, Zn and Se were analysed in the deposition modelling. The first one is based on the emission data for 2000 officially reported by Parties to the Convention to UNECE. For some of the countries, which do not report their national data, expert estimates of TNO (Denier van der Gon *et al.,* 2005) for 2000 were used. The second emission scenario assumes current legislation and current ratification (as of April 2005) of the HM Protocol. The third one implies that emissions will be controlled according to full implementation of the Protocol. The forth scenario envisages the conditions of the third one and a package of possible additional measures to control emissions. More details about emission projections for 2020 can be found in Denier van der Gon *et al.* (2005) and chapter 2.1 of this report. Further in the text, the results for 2020 scenarios "Current LEgislation", "Full Implementation of the Protocol" and "Full Implementation of the Protocol plus Additional Measures" are abbreviated as "CLE", "FI"or sometimes "FULL", and "FIAM", respectively.

Preparatory work of MSC-E to the review of MSCE-HM model has shown that the modelled concentrations and depositions of lead and cadmium based on officially reported emissions lead to a significant (2 – 3 times) underestimation of the observed values (Ilyin and Travnikov, 2005). Travnikov and Ilyin (2005) have also demonstrated that emission data are the major source of uncertainties of model estimates. The issue of emission data quality has been raised and discussed at meetings under the Convention. In particular, the Executive Body to the Convention "expressed concern at the insufficient reporting of emission data on heavy metals and urged Parties to work to improve this" (ECE/EB.AIR/87). The meeting of Task Force on Measurements and Modelling (TFMM) (Helsinki, Finland, 2006) also recognized that the significant difficulties with official emission data remain and further work to improve national emission estimates is needed. In this connection, in addition to Official/TNO emission data for 2000, the emission estimates for Pb, Cd, As, Ni and Cr produced within the ESPREME project (http://espreme.ier.uni-stuttgart.de/data.html) were used. The use of this emission data set for supporting an effect-based approach was recommended by TFMM at the meeting in Helsinki. Table 2.2.2 presents total emissions from Europe derived from the emission data sets involved in modelling. Table 2.2.2, shows that natural and historical emissions contribute a significant fraction to total emissions of heavy metals over Europe, according to current parameterization. The exception is cadmium, for which natural and historical emissions are several times lower than anthropogenic ones.

***Table 2.2.2*** *Total emissions from Europe in 2000, t/y*

| | Pb | Cd | Hg | As | Ni | Cr | Zn | Cu | Se |
|---|---|---|---|---|---|---|---|---|---|
| Official/TNO, 2000 | 11180 | 280 | 260 | 440 | 3840 | 1780 | 16700 | 2490 | 420 |
| 2020 CLE | 7900 | 260 | 305 | 360 | 3030 | 2090 | 16300 | 2240 | 310 |
| 2020 FI | 6200 | 190 | 300 | 270 | 2410 | 790 | 12600 | 1800 | 280 |
| 2020 FIAM** | 2300 | 115 | 165 | 150 | 800 | 360 | 9400 | 1500 | 170 |
| ESPREME, 2000 | 13160 | 580 | - | 760 | 4800 | 2700 | - | - | - |
| Natural and historical* | 6400 | 65 | 115 | 1040 | 990 | 1450 | 3570 | 1410 | 32 |

* Includes emissions from seas surrounding Europe: the North, Baltic, Mediterranean, Black and Caspian Seas.
**FIAM emissions have been used in the assessments and dispersion. These emissions imply an overestimate of the potential of additional measures of about 13 percent-points of updated emissions (see Table 2.1.4) , providing a wider range of depositions and exceedances between FI and FIAM.

Comparisons between emission data for 2000 and scenarios for 2020 require caution. For example, Table 2.2.2. shows that emissions of mercury and chromium in 2020 are higher than those for 2000. The reason for this is connected with the use of official emissions in 2000 for countries where they are available. Mercury emissions in Russia and some other countries in 2000 (official data) are lower than TNO estimates for 2020 by an order of magnitude (Figure 2.2.6a). Similar to mercury, official emissions of chromium for 2000 in some countries (e.g., France, Russia, Greece) are lower than TNO estimates for 2020 (Fig 2.2.6b). This led to higher total European emissions for 2020-CLE compared to totals computed for 2000.

The spatial distribution of emissions determines to a large extent the spatial pattern of depositions. As a rule, in regions where emissions are high, the depositions are also high compared to regions with low emissions. The spatial distribution of the sum of anthropogenic, natural and historical emissions in 2000, based on official/TNO and ESPREME data is given for Cd as example (Figure 2.2.7). ESPREME emissions of Cd are higher for most of countries of Europe compared to Official/TNO emissions (Figure 2.2.8). The obvious exceptions are some regions in Russia, Spain, Poland, Caucasus countries and some Balkan countries. From the viewpoint of modelling it is important to also note that not only total values for countries, but also the spatial distribution of these two data sets differs.

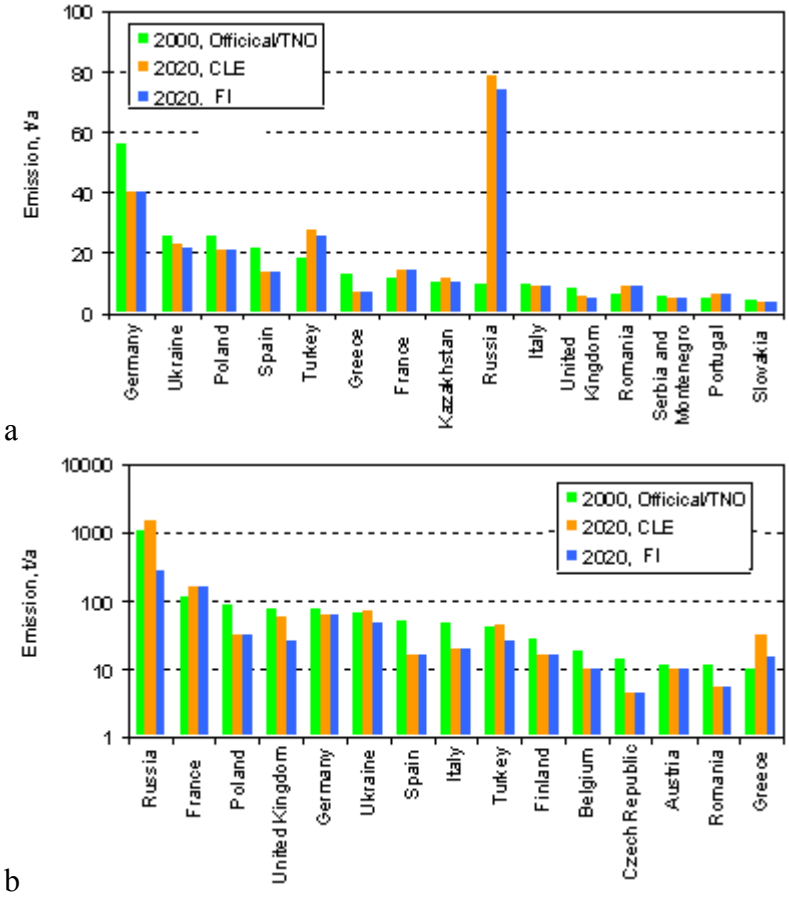


***Figure 2.2.6*** *Emission total values, t/a, of mercury (a) and chromium (b, logarithmic scale) in first 15 largest countries-emitters*

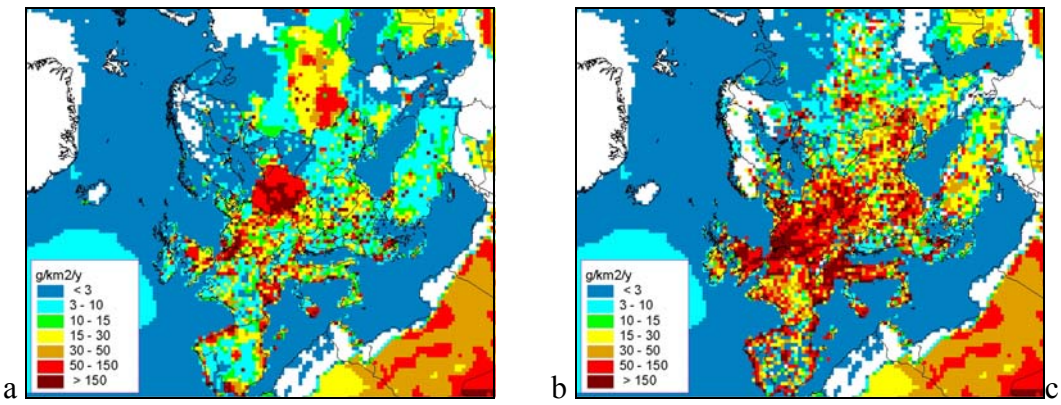


***Figure 2.2.7*** *Spatial distributions of emissions of cadmium in 2000 on the base of Official/TNO(a) and ESPREME (b) data. Natural and historic emissions included.*

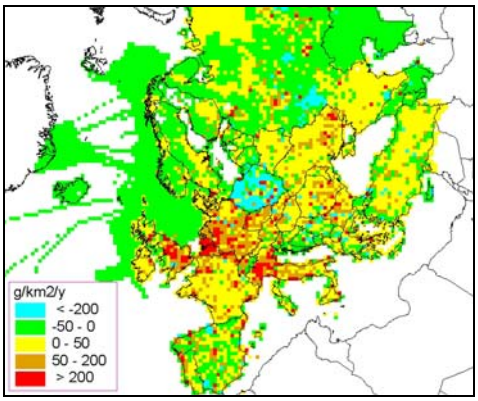


***Figure 2.2.8*** *Difference between ESPREME and Official/TNO emissions of cadmium in 2000*

## 2.2.3 Modelling results

Depositions of nine heavy metals (Cd, Hg, Pb, Cr, Ni, Cu, Zn, As, Se) have been calculated on the basis of emission scenarios for 2000 (combination of official data and TNO expert estimates) and "CLE", "FI" and "FIAM" scenarios 2020 (TNO expert estimates). In addition, ESPREME emissions have tentatively been used to compute ecosystem specific depositions of Cr, Ni, As, Cd, Pb in 2000. This section presents the results of deposition modelling of these metals. In particular, this section deals with spatial distributions of ecosystem-dependent depositions, and verification of modelling results via comparison with measurement data. According to the work plan of EMEP for 2006, MSC-E is responsible for pilot calculations of As, Ni and Cr concentrations and depositions. Furthermore, Cu, Cr and Se have not been included in the working plans before. Therefore, modelled depositions of metals not targeted by the Protocol (i.e, metals except for Pb, Cd and Hg) should be considered as preliminary.

*2.2.3.1 Spatial distribution of depositions*

The depositions were calculated for 18 types of land-cover. The computed depositions to all land-cover types as well as maps of mercury concentrations in precipitation have been transferred to the CCE, that used them to compute exceedances of critical loads for all land cover types (Average Accumulated Eceedance). However, to limit the number of maps in this chapter, the description of the deposition results in each EMEP grid cell focuses on crops and coniferous forests.

Examples of maps of depositions of cadmium in 2000 to coniferous forests and to crops, modelled on the base of Official/TNO emissions, is demonstrated in Figure 2.2.9. Over the major part of Europe the depositions to coniferous forests range from 0.15 to 1.5 g/ha/y (Figure 2.2.9a). Over the northern part of Russia the depositions are lower than 0.1 g/ha/y, and over the Scandinavian Peninsula they are below 0.03 g/ha/y. In Poland, the north-west of Germany, the east of Ukraine and some Balkan countries the depositions exceed 1.5 g/ha/y. Higher depositions in these regions are mainly caused by high emissions (anthropogenic and/or natural and historic ones) and less by surface roughness. Depositions to crops are lower (low deposition velocity of particles) than those to forested areas (Travnikov and Ilyin, 2005). Depositions to crops vary from 0.1 to 0.5 g/ha/y (Figure 2.2.9b) over most of Europe. Over the Scandinavian Peninsula, the north of Russia as well as most of France, Spain and Ireland the depositions are below 0.1 g/ha/y.

General peculiarities of spatial distribution of heavy metal depositions are similar to those for cadmium. Elevated depositions of heavy metals occur in regions where total emission (anthropogenic and natural and historic) is relatively high. These regions are Poland, central

part of Russia, Balkans, Belgium, north-western part of Germany, east of Ukraine. Relatively low depositions are computed in the Scandinavian Peninsula. Comparison of depositions derived from Official/TNO emissions and ESPREME data shows that ESPREME-based depositions are higher than those based on Official/TNO data over most of Europe.

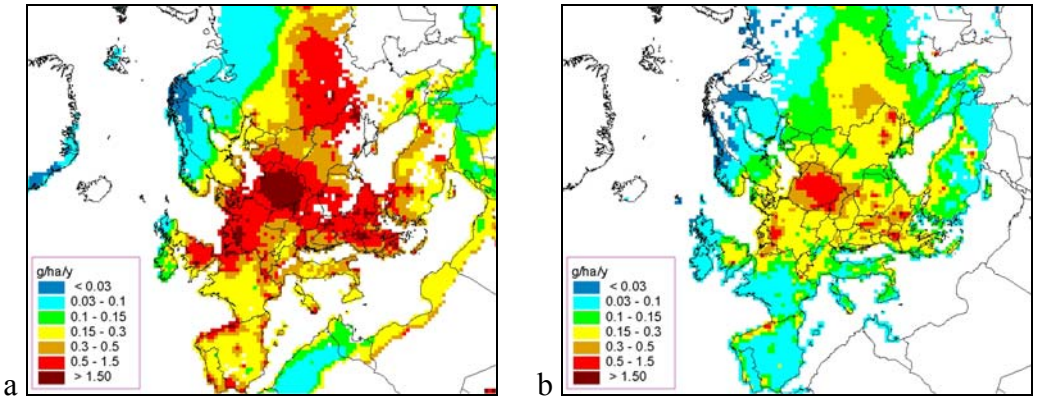


***Figure 2.2.9*** *Depositions of cadmium to coniferous forests (a) and crops (b) in 2000. Official/TNO emission scenario*

Land cover specific Cumulative Distribution Functions of depositions were compared to identify land cover characteristics. These functions demonstrate the cent of i-th land-cover area where depositions are below a certain value $d_i$. For example, modelled depositions below 0.3 g/ha/a occur over about 72% ($1.53 \cdot 10^6$ km$^2$) of coniferous forests and over 86% ($3.35 \cdot 10^6$ km$^2$) of croplands (Figure 2.2.10). Therefore, over 28% of coniferous forests and 14% of croplands the depositions exceed 0.3 g/ha/y.

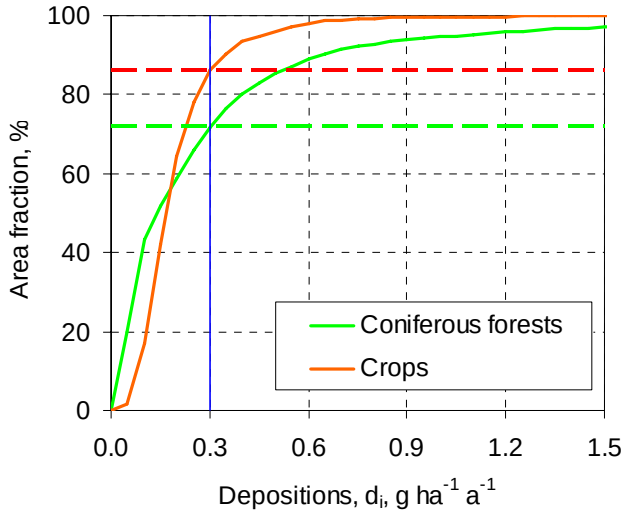


**Figure 2.2.10** *Cumulative distribution functions of cadmium depositions to coniferous forests and crops in 2000. Official/TNO emissions*

In order to calculate the effects of mercury on the environment, concentrations of mercury in precipitation are used as input parameter (UBA, 2004). Modelled concentrations of mercury in precipitation in Europe in 2000 vary from 7 to 20 ng/L (Figure 2.2.11). Higher concentrations were obtained for Germany, Poland, east of Ukraine, the Balkans. Similar to

pollution levels of other metals, these relatively high concentrations are associated with regions of significant mercury emissions.

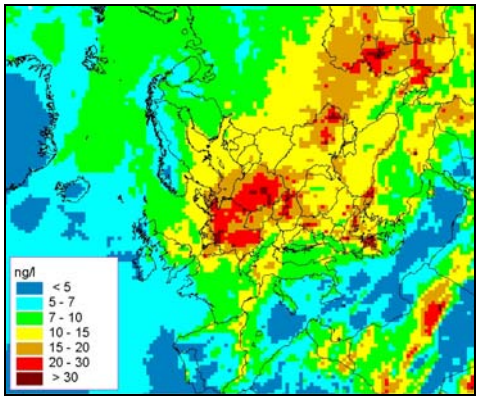


***Figure 2.2.11*** *Concentrations of mercury in precipitation in 2000. Official/TNO emission scenario*

*2.2.3.2 Evaluation of modelling results.*

Modelled depositions should be reliable for the robust evaluation of their effects on ecosystems. The real levels of pollution are determined by measurements. That is why the reliability of modelled data is verified via comparison of modelled parameters with measured quantities. In the ideal case the model should reproduce the observed values. In reality both the modelled and measured data contain some uncertainties. Besides, the model results are strongly depend on the emission input data. The available emission data sets for 2000 have been used to verify two parameters of the MSCE-HM model against measurements available from the Chemical Coordinating Centre, i.e. concentrations in air and concentrations in precipitation. An uncertainty analysis described in Ilyin and Dutchak (2005) revealed that reported emissions are too low to reflect depositions.

*2.2.3.3 Depositions of heavy metals in 2020*

Ecosystem-dependent depositions of the nine metals were calculated for three TNO emission projections for 2020. Examples shown in Figure 2.2.12 demonstrates total depositions of cadmium based on “CLE” and “FI” scenarios for 2020 in comparison with Official/TNO estimates for 2000. As seen, depositions of cadmium to Europe as a whole according to these two emission projection scenarios are going to decrease by 2020. This decrease is clearly seen, e.g., in Poland, Spain, or countries of Balkan Peninsula. As the modelling used the same meteorological data for 2000 and 2020, and the same natural and historic emissions, the difference in depositions is explained only by differences in emission data.

In some regions (e.g. Kola Peninsula in Russia, Ukraine) modelled depositions in 2020 are higher than those for 2000 as can be seen from Figure 2.2.13 that shows the ratio of cadmium depositions in 2000 to the depositions in 2020 (scenario “CLE”). The ratio below unity means a decrease of depositions between 2000 and 2020. The ratio may exceed 2. The reason for this is due to the derivation of emission data in 2000 and 2020. Emissions for 2000 are based on official data whereas emission data for 2020 are based on expert estimates of TNO (see chapter 2.1). Official emissions of cadmium (and other metals) in some countries, e.g. Russia, in 2000 are lower than expert estimates of TNO for 2020 which is reflected in an increase of deposition between 2000 and 2020 in some EMEP grid cells.

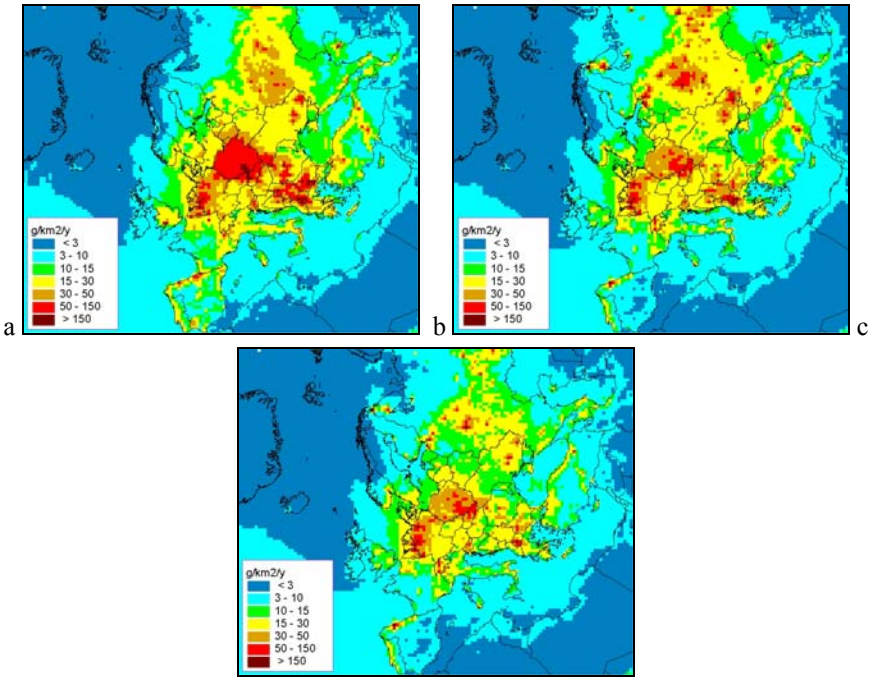


***Figure* 2.2.12** *Spatial distribution of total depositions of cadmium in 2000 and 2020. (a): 2000, official/TNO emissions; (b): 2020, "CLE"; and (c) 2020, "Full"*

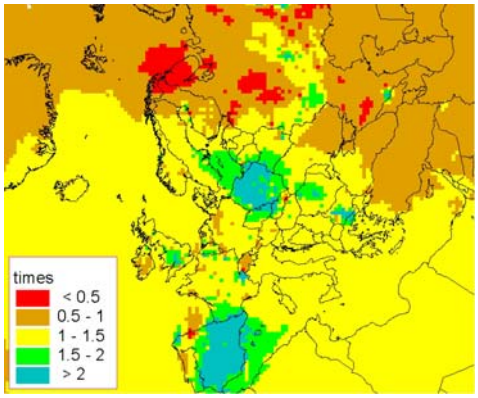


***Figure 2.2.13*** *Ratio of cadmium depositions in 2000 (official/TNO emissions) to depositions in 2020 (scenario "CLE").*

Similar to depositions of cadmium and other metals, the concentrations of mercury in precipitation in 2020 are lower than those in 2000 over most of Europe (Figure 2.2.14). A marked decrease can be noticed in Germany, Poland, Belgium, and the United Kingdom. However, Russia is an exception of this tendency because of differences in approaches to set their emission data. As shown in Figure 2.2.6, the estimates of mercury emissions in Russia in 2020 are by an order magnitude higher than the officially reported value for 2000.

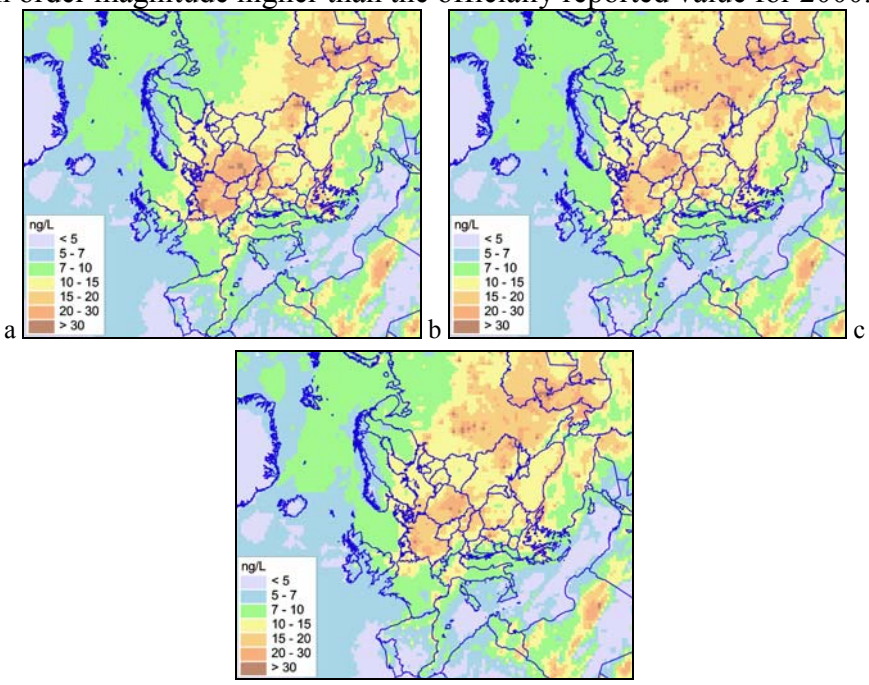


***Figure 2.2.14.*** *Spatial distribution of mercury concentrations in precipitation in 2000 and 2020. (a): 2000, official and TNO emissions; (b): 2020, "CLE"; and (c) 2020, "Full"*

The depositions for three scenarios for 2020 were calculated for all other metals which have been made available to the CCE for use in the assessment of exceedances (see chapter 3).

## 2.2.4. Concluding remarks

This section summarizes the main outcomes of the modelling of ecosystem-dependent depositions and mercury concentrations in precipitation that have been delivered by MSC-E to the CCE.

- Estimated magnitudes of natural and historic emissions of aerosol-bound heavy metals are comparable to anthropogenic emissions. As natural and historic emissions are a function of long-term heavy metal accumulation, the investigation of this accumulation requires further research.
- Annual mean concentrations in air and in precipitation of Pb, Cd, As, Ni and Cr based on ESPREME emission estimates are generally higher than those based on a combination of official data and TNO expert estimates.
- The results computed on the base of official/TNO emissions for 2000 underestimate measurements. The use of ESPREME emission estimates may result in some overestimation of measured quantities. In particular, concentrations of arsenic in the air, chromium in precipitation, and nickel concentrations in both media were somewhat overestimated. In general, the agreement between observed and modelled values based on ESPREME emissions is better.
- Modelled concentrations and depositions of Cd, Pb, Cu, Zn and Se are significantly lower than measured data. Exceedances that are based on these results should be considered as very preliminary and handled with care as they may be lower than exceedances that are computed based on measured data.
- Depositions in 2020 in some regions can be higher than those in 2000 by a factor of 2 or even more. This is due to the manner in which emissions in 2000 (official/TNO emissions) and 2020 (TNO assessment) were obtained. Caution should be taken when comparing depositions in 2000 and 2020. Further work is needed to improve the emission data.

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## 2.3 Critical Loads of cadmium, mercury and lead

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### 2.3.1. Summary

Critical loads of cadmium, mercury and lead have been produced in collaboration with National Focal Centres using methods which are described in UBA (2004). Results have been published in Slootweg et al. (2005a; 2005b). At the end of 2005 until the beginning of 2006 Parties had been enabled to submit updates of the critical load of heavy metals that had been adopted by the 24th meeting of the Working Group on Effects and published in CCE (2005). Altogether, 18 countries submitted critical loads of heavy metals. However, not all countries addressed each of the five effects that are described in Table 2.3.1.

***Table 2.3.1*** Overview of indicators used in the computation of critical thresholds. *(Source: Slootweg et al., 2005b)*

| *Receptor Ecosystem* | *Endpoints* | *Heavy metals of concern* | *Land cover types to be considered* | *Indicator/critical limit* | *Effect number* |
|---|---|---|---|---|---|
| *Terrestrial* | *Human health effects* | Cd, Pb, Hg | All ecosystems | Total concentration in soil water below the rooting zone (to protect ground water) | 1 |
| | | Cd, Pb, Hg | Arable | Content in food, fodder and crops | 2 |
| | | Cd, Pb, Hg | Grassland | Content in grass and animal products (cows, sheep) | |
| | *Ecosystem functions* | Cd, Pb | Arable land, grassland, non-agricultural | Free ion concentration in view of effects on soil micro-organisms, plants and invertebrates | 3 |
| | | Hg | Forest soils | Total concentration in humus layer in view of effects on soil micro organisms and invertebrates | |
| *Aquatic* | *Ecosystem functions* | Cd, Pb, Hg | Freshwaters | Total concentration in view of effects on algae, crustacea, worms, fish, top predators | 4 |
| | *Human health* | Hg | Freshwaters | Concentration in fish | 5 |

Table 2.3.2 gives an overview of the effects that were addressed by Parties. It shows that most countries computed critical loads for effects 1 and 3. In the beginning of 2006 updates have

been submitted by Bulgaria, Cyprus, Italy, The Netherlands and the Ukraine. Critical loads of cadmium, lead and mercury have been computed by 17, 17, and 12 countries, respectively, including results of the early 2006 update.

Posch and Reinds (in Slootweg et al, 2005a) combined location and receptor data to create a European 'background' database of critical loads, which could be used to compute critical loads for areas in countries that did not participate in the mapping exercise. Few countries have objected to the use of this dataset for their territory.

***Table 2.3.2*** *Overview of available national data on critical loads of cadmium, lead and mercury for the 5 effects (Based on: Slootweg et al., 2005b)*

| Country | Country Code | Effect number (Table 2.3.1) | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Cd | | | | Pb | | | | Hg** | | |
| | | 1 | 2 | 3 | 4 | 1 | 2 | 3 | 4 | 1 | 3 | 5 |
| Austria | AT | x | x | x | | x | | x | | x | x | |
| Belarus | BY | | | x | | | | x | | | | |
| Belgium | BE | x | | x | x | x | | x | x | x | x | x |
| Bulgaria | BG | x | | | | x | | | | | | |
| Cyprus | CY | x | x | x | | x | | x | | x | | |
| Czech Republic | CZ | x | | | | x | | | | x | | |
| Finland | FI | | | | | | | | | | | x |
| France | FR | | | x | | | | x | | | | |
| Germany | DE | x | x | x | | x | | x | | x | x | |
| Italy | IT | | | x | | | | x | | | | |
| Netherlands | NL | x | x | x | | x | | x | | | x* | |
| Poland | PL | | | x | | | | x | | | x | |
| Russia | RU | x | | x | | x | | x | | | | |
| Slovakia | SK | | | x | | | | x | | | x | |
| Sweden | SE | | x | x | | | | x | | | x | x |
| Switzerland | CH | x | | x | | x | | x | | | x | |
| Ukraine | UA | | x | | | | x | | | x* | | |
| United Kingdom | GB | | | x | | | | x | | | | |
| Total | 18 | 10 | 6 | 14 | 1 | 10 | 1 | 14 | 1 | 7 | 8 | 3 |

* Updated in 2006. This Effect was not part of the 2005 data submission.
** Belgium, Sweden and Finland also assessed critical concentrations in precipitation

The critical loads that have been submitted by the Parties are used in the following of this report to compute the exceedances in 2000 and 2020 following CLE, FI and FIAM.

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## 2.4 Critical Loads of copper, nickel, zinc, arsenic, chromium and selenium for terrestrial ecosystems at a European scale: A preliminary assessment

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### 2.4.1 Methods

The method to calculate critical loads of Cr, Ni, Cu, Zn , As and Se is based on the balance of all relevant metal fluxes in and out of a considered ecosystem in a steady state situation and is identical to the method for Cd and Pb described in the mapping manual (UBA, 2004).

*Model*

The critical load of a metal can be calculated from the sum of tolerable outputs from the considered system in terms of net metal uptake and metal leaching. The critical load equals the net uptake by forest growth or agricultural products plus an acceptable metal leaching rate, according to:

$$CL(M) = M_u + M_{le(crit)} \quad (1)$$

where:

$CL(M)$ = critical load of a heavy metal M ($g.ha^{-1}.yr^{-1}$)

$M_u$ = Metal net uptake in harvestable parts of plants under critical load conditions ($g.ha^{-1}.yr^{-1}$)

$M_{le(crit)}$ = critical leaching flux of heavy metal M from the considered soil layer ($g.ha^{-1}.yr^{-1}$), whereby only the vertical drainage flux is considered

The critical leaching flux of a heavy metal from the topsoil can be calculated by multiplying the flux of drainage water with the critical total concentration of heavy metal in soil drainage water.

The removal of metals by net growth and harvest at the critical load refers to the steady state level at critical load. For many metals however there is no clear relationship between concentrations in soil solution and concentrations in the harvestable parts in plants (both agricultural crops and trees). We have calculated the uptake from an average metal content in plants as found in relatively unpolluted areas. Because site specific data on metal contents are not available at a European scale we use default values for metal contents in trees and crops. Details on the procedure and element contents used are provided in Reinds et al. (2006).

*Critical limits*

Critical concentrations of the metals As, Cr, Cu, Ni, Se and Zn in soil drainage water, depend on the target to be protected and can be derived as:

- Concentrations of free metal ions or total concentrations in soil solution (As, Cr, Cu, Ni, Se and Zn) in view of ecotoxicological effects on soil micro-organisms, plants and invertebrates.

- Metal concentrations (total dissolved concentration) in ground water in view of human health effects through intake of drinking water.

For zinc the critical metal contents in plants in view of animal health effects through intake of plant products was used to make a first example calculation of a critical load for animal health (Reinds, et al., 2006). For other metals such limits are not available.

The difference in toxicity between the metals is reflected in the differences in critical concentrations as shown in Table 2.4.1:

*Table 2.4.1: Critical limits for concentrations of As,Cr,Cu,Ni,Se and Zn related to drinking water quality and eco-toxicological effects*

| | *Critical concentration in µg.l$^{-1}$* | |
|---|---|---|
| *Metal* | *Human health, drinking water* | *Eco-toxicology* |
| Cr | 50 | 44 |
| Ni | 20 | ± 25 – 700 (pH and DOC dependent function) |
| Cu | 2000 | ± 1 – 50 (pH and DOC dependent function) |
| Zn | - | ± 20 - 90 (pH and DOC dependent function) |
| As | 10 | 70 |
| Se | 10 | 1 |

Table 2.4.1 shows for example that the drinking water limit for arsenic, that is considered to be rather toxic, is lower than the limit for copper. Low critical concentrations lead to relatively low critical loads. Therefore, if a tentatively ranking of human health toxicity by critical concentration is considered everything else being equal, then arsenic and selenium come out as more toxic than nickel, chromium and copper. When, under these conditions, the minimum is taken of public health and eco-toxicological related critical concentrations, the ranking becomes selenium, copper, arsenic, nickel, zinc and chromium. However, specific environmental conditions also contribute to the magnitude of a critical load.
Also note, that zinc is an essential element for all living organisms. Therefore, no drinking water limit exists for zinc. Critical concentrations for Cu, Ni and Zn related to eco-toxicological effects were computed as a function of soil pH and DOC in accordance with the method described in the Mapping Manual (UBA, 2004) for Cd and Pb. Unfortunately, such functions are not available for As, Cr and Se. Eco-toxicological limits for these elements were derived from few literature data and should thus be considered to be a first estimate (Reinds et al., 2006).

## 2.4.2 Input data

Geographical input data for the critical load model include precipitation surplus, soil organic matter content, soil pH, DOC concentration in the top soil, forest growth and crop yield. These input data vary as a function of location and/or ecosystem type.

Maps with computational units (receptors) that hold the required information to derive the input data for the model were constructed by overlaying maps with a grid resolution of 2.5 × 2.5 km. Two map overlays were made, one that holds the spatial distribution of receptors for forests[1] and one for agricultural soils.

[1] Forestry data on growth are missing for Belarus and parts of the Ukraine and the Southern part of Russia.

In the following a brief overview is given on how the input parameters were derived from existing databases; details are provided in Reinds et al. (2006).

Precipitation surplus was derived using a simple hydrological model that was applied to each receptor. Meteorological input data for the period 1961-1990 were obtained from a detailed meteorological database that provides high resolution data for the period 1900-2000.

Metal uptake was estimated from yield data and metal contents in crops and wood. Yield data for agricultural crops were obtained from EU data sets limiting the area for which critical loads could be mapped to the EU countries, whereas stemwood increment was obtained from a data set that provides national forest growth data for about 120 regions in Pan-Europe.

Topsoil pH was derived from soil profile databases, distinguishing between a database for forest soils to estimate forest soil pH and a database targeted towards agricultural soils. pH data were related to soil maps based on soil type. Dissolved organic carbon was computed using transfer functions that estimate DOC concentrations as a function of pH and organic matter.

### 2.4.3 Results

Maps showing the spatial distribution of critical loads over EMEP grid cells are presented in chapter 3 for each metal separately. For each EMEP 50×50 km cell the $5^{th}$ percentile critical load is illustrated. The $5^{th}$ percentile critical load is the critical load value below which 5 percent of the critical loads lie. This implies that a deposition value that is equal to the $5^{th}$ percentile critical load will protect 95 % of the ecosystems in the grid cell. Maps are shown for either ecotoxicological effects or for human health effects through drinking water, depending on which of two effects yielded the lowest critical load. Ecotoxicology related critical loads are shown for forest areas, human health related critical loads for agricultural areas.

### 2.4.4 Conclusions

Because of its intrinsic simplicity, the critical load concept could also be applied to the metals Cu, Zn, Ni, Cr, As and Se. For Cu, Zn and Ni, critical load functions could be derived that provide critical concentrations related to ecotoxicological effects as a function of pH and DOC. For the other metals only a fixed total concentration was used, independent of pH and DOC. It is likely however, that for the cationic metals the toxic effects are better related to free metal ions, so critical total concentrations also depends on pH and DOC. For more accurate assessments of critical concentrations, it should be investigated if critical load functions can be derived for these metals as well. As and Se are present in soil solution as anions. Possibly other factors play a role in the toxicity of these elements than for the cationic metals.

Results show that for most metals and receptors investigated, leaching is the dominant term in the critical load. As a consequence, both the critical metal concentration and the leaching flux are important parameters. Leaching was estimated from detailed rainfall data and modelled water consumption using a simple water balance model. Modelled leaching fluxes are uncertain, as no calibration or validation of the model has been carried out at the European

scale. However, it has been shown that even such simple models can perform rather well when looking at the plausibility of the simulated leaching using chloride budgets at Intensive Monitoring plots (De Vries et al., 2003).

Most of the uncertainty in the critical loads for ecosystem protection stems from the uncertainty in the critical concentrations used. Especially for Cr, As en Se, critical concentrations stem from very few sources and are therefore highly uncertain. For more robust assessments of critical metal loads for ecosystem protection, a through review of existing ecotoxicological data for these elements is needed.

Experimental data indicate that the concentration of chromium in soil solution is determined by the solubility product of the hydroxide and hence there will also not be a steady state concentration related to the input flux of Cr to soil. This is in contradiction with the steady state assumption of the critical load approach which means that if Cr concentrations in soil solution are indeed regulated by the solubility of a chromium precipitate, the calculation of a critical load for Cr by the current methodology is invalid.

## References

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UBA, 2004. Mapping manual 2004. Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. www.icpmapping.org.

## 2.5 Exceedance of Critical Loads and uncertainty

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Exceedances are computed by comparing critical loads and levels with atmospheric depositions. Depending on the endpoint, exceedances can be computed to identify the risk for human health or the risk for eco-toxicological effects (see chapter 2.3). All the critical load data are used to compute the Accumulated Average Exceedance (AAE). If both endpoints are applicable to any single ecosystem point then three kinds of AAE can be computed, i.e. AAE with respect to human health, AAE with respect to eco-toxicology and the AAE with trespect to the minimum critical load of both end points. Results are illustrated for cadmium, lead, mercury, chromium, nickel, copper, zinc, arsenic and selenium in paragraphs 3.1.4, 3.2.4 until 3.9.4 respectively.

### 2.5.1 Accumulated Average Exceedance[2]

Accumulated Average Exceedances (AAE) have been computed to identify and map areas (grid cells) where atmospheric metal depositions are higher than critical loads. An AAE is the ecosystem area-weighted sum of the individual exceedances (deposition minus critical load, with zero for non-exceedance) of all ecosystems in a grid cell. The AAE is defined as:

$$AAE = (A_1Ex_1 + \ldots + A_nEx_n)/(A_1 + \ldots + A_n)$$

where $A_i$ is the area of the $i$-th ecosystem in a grid cell and $Ex_i$ its exceedance ($i$=1,…,$n$). In Figure 2.4.1 the AAE for a given deposition ($D_a$) is given by the grey area. (See also Posch et al., 2001; UBA, 2004).

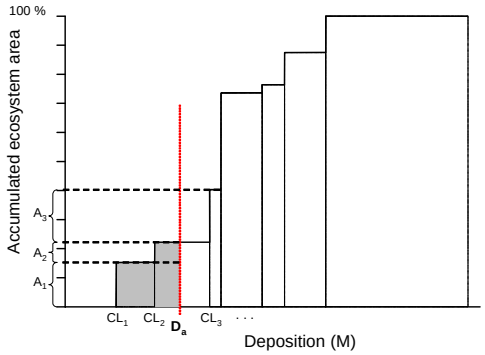


***Figure 2.5.1*** *Accumulated Average Exceedance (AAE).*

The AAE has been computed with respect to health, ecosystem and combined effects. The latter computes the AAE by using all data on critical loads irrespective of whether the endpoint is human or ecosystem health. The minimum critical load is taken if for an area critical loads are available for both end points.

[2] The text of this section draws upon Slootweg *et al.* (2005), p.61.

The AAE has been computed for European ecosystem areas using ecosystem specific deposition of heavy metals and critical loads of European ecosystem as described in section 2.2 and 2.3 respectively within each EMEP grid cell. Thus the AAE is computed for 2000, 2020 Current Legislation (CLE), 2020 Full Implementation (FI) and 2020 Full Implementation plus Additional Measures (FIAM). The exceedances that are computed under the FIAM scenario are an underestimate. This is caused by a difference in FIAM emissions used in the dispersion model and updated TNO data (see Tables 2.1.4 and 2.2.2) at the end of the project. The result of this is that the estimate of the "actual" risk for adverse effects of FIAM – as presented in the exceedance sections in chapter 3 - should be interpreted in the range of the reported FI and FIAM exceedances. This can lead to the following cases. If estimated protection (of the European ecosystem area) is already achieved under FI than FIAM does not add any effect-based information. If protection is not yet achieved under FIAM, this implies that the use of latest updated information on FIAM emissions (which are higher) would further increase the risk. Finally, if estimated protection is not achieved under FI but realized under FIAM then further assessment of the risk using updated FIAM emissions may provide improved information on the requirements to achieve protection, if appropriate.

### 2.5.2 Uncertainty

The uncertainty of critical loads is only one of the elements contributing to reliability of the critical load approach. Error propagation due to emission data uncertainties, and uncertainties in atmospheric transport as well as critical load modelling, require a cautious treatment of quantitative results, whereby emission data are used in dispersion models to compute depositions that are finally compared to critical loads to produce exceedances. Therefore, the use by means of scenario analysis of quantitative results under the LRTAP Convention focuses on relative, rather than on absolute assessments. When comparing the effects of different scenarios of emission changes, the variability between scenarios is primarily due to the variation in policy options in each scenario, rather than due to error propagation within each scenario. Methodological causes of error propagation can be assumed to be fairly similar independent of the scenario (See also Hettelingh and Posch, 1997). Therefore, it is recommended to interpret the scenario-specific (i.e. CLE, FI, FIAM) risks of adverse effects (exceedances) – as presented in chapter 3 - in comparison to 2000 and to each other.

In TFHM (2006) an overview of the knowledge on uncertainty of the critical load approach is provided, which is not repeated here. With respect to the report before you it is acknowledged that scientific review of the critical load assessments of cadmium, mercury and lead is more extensive than for the 6 other metals. While this is especially true for the derivation of critical limits, it is noted that the methodology is similar to that used for the assessment of critical loads for cadmium, mercury and lead. The choice and derivation of the critical limits is summarized in this report, while for details the reader is referred to the background document (Reinds et al., 2006).

The critical loads approach for heavy metals, including methods to derive critical limits, is the result of careful review of internationally accepted methods. Its scientific basis represents the state of knowledge. While uncertainty of emissions and depositions can be reduced by verification and validation procedures, this is more difficult for critical loads.
Oreskes et al. (1994) explaines why verification and validation of numerical models describing open natural systems is impossible.

Along these lines one can argue that validation of critical loads is very challenging, if anything because it may take until a future generation before the appropriateness of the critical load as a risk indicator is established, i.e. before critical load exceedance leads to actual damage. However, verification, i.e. the comparison of model behaviour to expectations, is possible to a certain extent and is an important element of future work. Some uncertainties can be reduced as more or better information becomes available.

## References


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Posch M, Hettelingh J-P, De Smet PAM (2001) Characterization of critical load exceedances in Europe. *Water, Air and Soil Pollution* **130**: 1139-1144

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UBA (2004), Manual on methodologies and criteria for modelling and mapping of critical loads and levels and air pollution effects, risks and trends, www.icpmapping.org.

# 3. Results[3]

This chapter presents a summary of the results for each of the 9 heavy metals metals, mostly expressed as tables or maps, of (1) emissions, (2) depositions, (3) critical loads and finally (4) exceedances of each of the nine heavy metals.

A recent overview of the current effects of heavy metals with emphasis on the 3 priority metals has recently been compiled in the Sufficiency and Effectiveness reports under the Task Force on Heavy Metals (TFHM, 2006a; 2006b).

The results in this report, in particular regarding emissions and exceedances, focus on 2000 emission and three scenarios for 2020, i.e. "Base Line scenario with Current Legislation and Current Ratification of the HM Protocol" (CLE), "Base Line scenario with Current Legislation and Full Implementation of HM Protocol" (FI); and "Current Legislation and Full Implementation of the HM Protocol plus possible Additional Measures" (FIAM). Emissions are computed for 2000 as base year, for 2010 and 2020 under CLE, and for 2020 under both FI and FIAM.

The first three sections focus on the three priority metals, i.e. cadmium, mercury and lead. Sections 4-9 describe results regarding chromium, nickel, copper, zinc, arsenic and selenium respectively.

Exceedance results include (a) a table of country specific percentages of ecosystem area at risk which enables the user-calculation of $km^2$ at risk, and (b) maps showing an interval of the magnitude in g $ha^{-1}yr^{-1}$ of the Accumulated Average Exceedance for each endpoint, i.e. human health, ecosystem health and a combination of the two.
The maps are structured on one page as a grid of 9 maps, i.e. 4 rows of 3 maps. The columns distinguish mapped results with respect to the endpoints (3 columns of maps). The rows distinguish between results in 2000, 2020-CLE, 2020-FI and 2020-FIAM (4 rows).

[3] The sections on emissions are by Hugo Denier van der Gon, Maarten van het Bolscher and Antoon Visschedijk (TNO), the sections on depositions by Ilia Ilyin and Oleg Travnikov (EMEP/MSCE) and the sections on critical loads are by Gert Jan Reinds, Bert Jan Groenenberg and Wim de Vries (ALTERRA), except for cadmium, lead and mercury which are from the CCE. Finally, the sections on critical load exceedances are by Jean-Paul Hettelingh and Jaap Slootweg (CCE).

# 3.1 Cadmium

## 3.1.1 Emissions

***Table 3.1.1*** *National cadmium emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 199 | 196 | 204 | 190 | 162 |
| Armenia | | 129 | 156 | 147 | 142 | 141 |
| Austria | | 1427 | 1442 | 1432 | 1432 | 1228 |
| Azerbaijan | | 2291 | 2767 | 2712 | 2702 | 313 |
| Belarus | | 1394 | 1489 | 1417 | 906 | 591 |
| Belgium | | 2750 | 1566 | 1583 | 1583 | 922 |
| Bosnia-Herzegovina | | 1675 | 1572 | 1423 | 602 | 430 |
| Bulgaria | | 10986 | 5409 | 4244 | 4244 | 2172 |
| Croatia | | 1019 | 815 | 666 | 601 | 435 |
| Cyprus | | 572 | 612 | 725 | 725 | 71 |
| Czech Republic | | 2840 | 1134 | 991 | 991 | 846 |
| Denmark | | 986 | 734 | 701 | 701 | 668 |
| Estonia | | 680 | 322 | 198 | 198 | 134 |
| Finland | | 1400 | 1353 | 1394 | 1394 | 990 |
| France | | 10453 | 9056 | 7859 | 7859 | 5588 |
| Georgia | | 210 | 265 | 273 | 262 | 139 |
| Germany | | 21062 | 20156 | 21571 | 21571 | 17879 |
| Greece | | 2844 | 2378 | 2249 | 2249 | 1293 |
| Hungary | | 2748 | 2376 | 2563 | 2563 | 2328 |
| Iceland | | 81 | 85 | 84 | 84 | 81 |
| Ireland | | 1341 | 886 | 764 | 764 | 586 |
| Italy | | 11051 | 6927 | 6607 | 6607 | 4405 |
| Kazakhstan | | 19239 | 22386 | 23978 | 14410 | 6738 |
| Kyrgyzstan | | 347 | 433 | 393 | 307 | 294 |
| Latvia | | 589 | 350 | 366 | 366 | 182 |
| Lithuania | | 1367 | 1610 | 1488 | 1488 | 1383 |
| Luxembourg | | 51 | 55 | 57 | 57 | 53 |
| Macedonia (Form. Yug. Rep. of) | | 9764 | 9623 | 9461 | 4582 | 1291 |
| Moldova (Rep. of) | | 373 | 332 | 324 | 324 | 265 |
| Netherlands | | 1158 | 1118 | 1174 | 1174 | 861 |
| Norway | | 725 | 936 | 975 | 975 | 781 |
| Poland | | 50400 | 29481 | 20627 | 20627 | 18792 |
| Portugal | | 3241 | 2350 | 2472 | 2472 | 1587 |
| Romania | | 17368 | 4274 | 4432 | 4432 | 1457 |
| Russia | | 111514 | 123849 | 131744 | 62518 | 28279 |
| Slovak Republic | | 7248 | 3321 | 3168 | 3168 | 2860 |
| Slovenia | | 1542 | 1320 | 1415 | 1415 | 1265 |
| Spain | | 15521 | 9856 | 6850 | 6850 | 5858 |
| Sweden | | 425 | 373 | 329 | 329 | 243 |
| Switzerland | | 2176 | 2892 | 3037 | 3037 | 2761 |
| Turkey | | 16640 | 17915 | 18853 | 9804 | 6283 |
| Ukraine | | 23682 | 19093 | 19546 | 12698 | 9350 |
| United Kingdom | | 7249 | 4886 | 4691 | 4691 | 3615 |
| Yugoslavia (Fed. Rep. of) | | 8653 | 8426 | 7809 | 2623 | 1422 |
| ***Total (tonnes/yr)*** | | ***377*** | ***327*** | ***323*** | ***217*** | ***137*** |

## 3.1.2 Depositions

Depositions for Cd are calculated for the 2000 official/TNO emissions and the three emission scenario's for 2020, all including natural and historic emissions. The calculated Cd depositions are most probably an underestimation.

Concentrations of cadmium in air and in precipitation based on official/TNO emission data for 2000 are significantly (a factor 2 - 3) underestimated (Fig 3.1.1). Correlation coefficients were significant: 0.76 for concentrations in air, and 0.84 in precipitation. These coefficients indicate that the spatial distribution of pollution levels is correctly reproduced by the model, but magnitudes of official/TNO emissions or natural and historic emissions seem considerably underestimated. This underestimation means that real atmospheric load to the ecosystems is significantly higher than that produced by modelling on the base of official/TNO emissions. This fact should be taken into account when comparing modelled atmospheric loads with their threshold values.

The use of ESPREME emissions leads to much better agreement between modelled and measured quantities for Europe as a whole: the underestimation for concentrations is quite small (~10%) and for concentrations in precipitation – about 30%. These values are similar to the uncertainties of MSCE-HM model [Travnikov and Ilyin, 2005]. Higher modelled parameters are explained by higher emissions of ESPREME compared to official/TNO data. However, correlation coefficients for ESPREME-based concentrations in air and in precipitation are relatively low: 0.56 and 0.53, respectively. It means that at some measurement stations the modelled results may significantly deviate from the observed values.

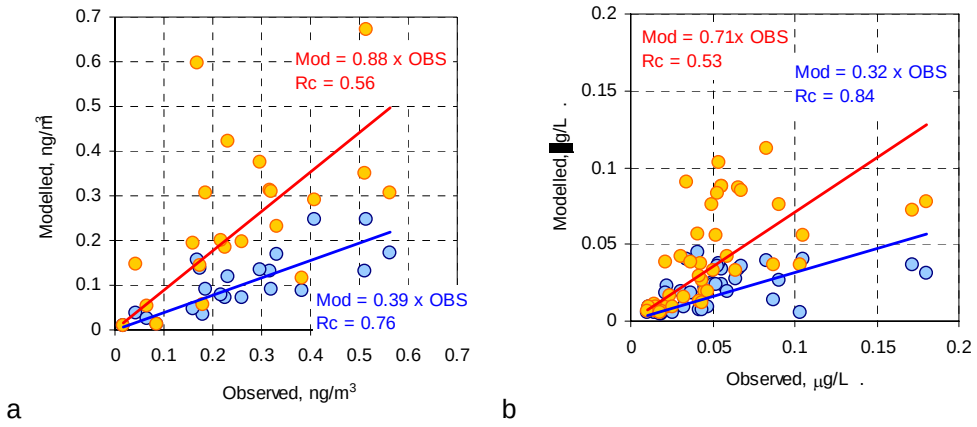



***Figure 3.1.1*** *Comparison of modelled and measured cadmium concentrations in air (a) and concentrations in precipitation (b) using official/ TNO (blue) and ESPREME (red) emissions. Natural and historical emissions are included. Rc –correlation coefficient*

Comparison of modelled air concentrations of cadmium, computed on the basis of ESPREME and natural and historical emissions, against measurements at individual stations is demonstrated in Figure 3.1.2. For bars indicating modelled values the contributions of anthropogenic emissions, natural and historical emissions and background concentrations are marked. The modelled concentrations are mainly determined by the anthropogenic component. The contribution of natural and historical emissions at most of stations does not exceed 13%, and background concentrations – 6%. At some of stations, e.g. the Dutch and British the model considerably overpredicts the observed values. Moreover, even the use of anthropogenic emission only would result in the overprediction. The ESPREME emissions in

the United Kingdom are as much 5 times larger than those of official/TNO. For the Netherlands, and neighbouring Belgium and Germany the ESPREME emissions are larger 9, 6 and 3 times, respectively. Therefore, these emissions may be too large, resulting to the overestimation of observed concentrations by the model. Besides, the comparison of modelled air concentrations based on official/TNO plus natural and historical emissions shows that for stations NL9 and GB91 the model agrees well with measurements (Figure 3.1.3.). On some other stations (e.g., located in Latvia, Slovakia) the situation is opposite: despite the use of relatively high ESPREME emissions and natural and historical emissions the observed concentrations are significantly (up to three times) underestimated. The reasons of the discrepancies between the model and measurements can be connected with uncertainties of emission magnitude and its spatial allocation, and uncertainties of the model parameterisations. More detailed investigation of these reasons is needed.

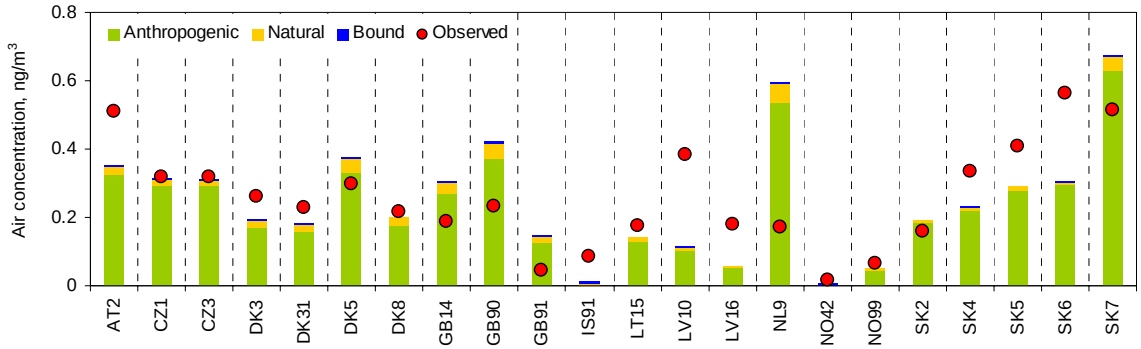



***Figure 3.1.2*** *Contribution of anthropogenic emissions (ESPREME estimates), natural emission and boundary concentrations to modelled air concentrations of cadmium*

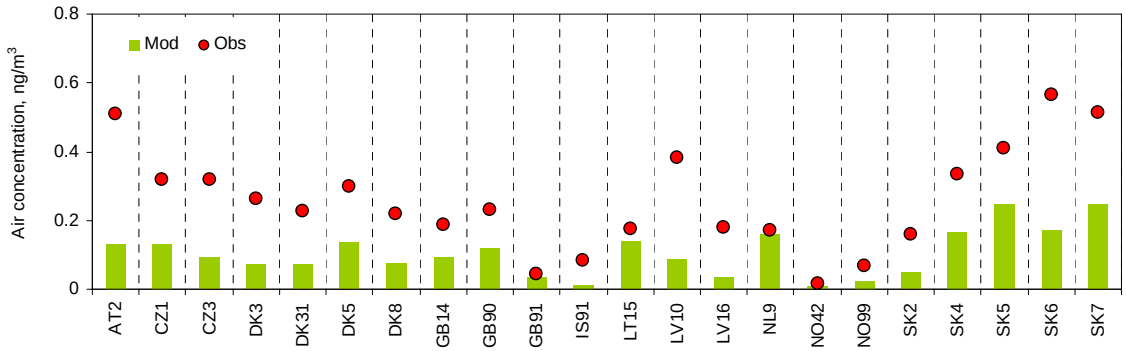
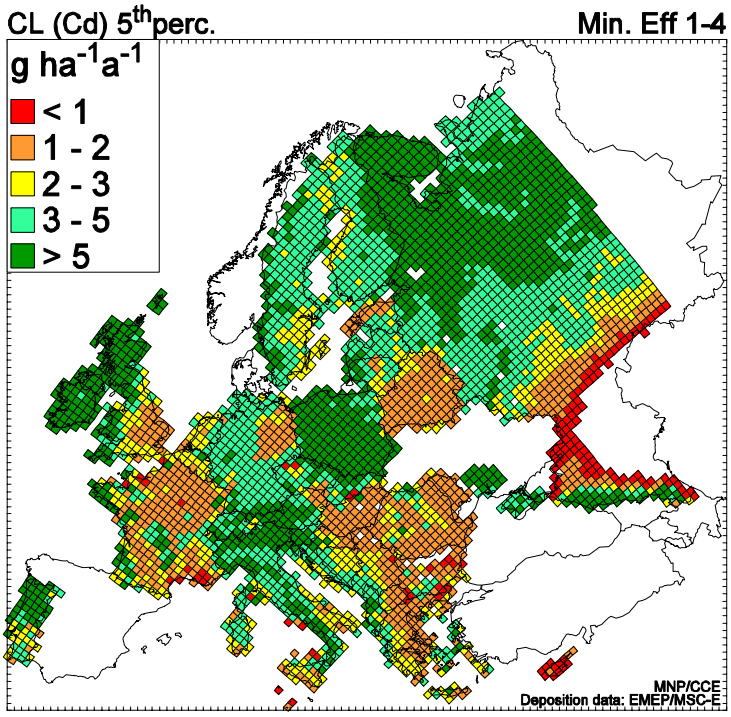



***Figure 3.1.3*** *Comparison of modelled and measured concentrations of cadmium in air. Modelling results are based on official/TNO emissions including natural and historical emission.*

## 3.1.3 Critical loads of cadmium

Computed European critical loads that are estimated to protect 95% of the ecosystems from the risk of both human and ecological health effects are shown in Figure 3.1.4. The CCE background database was used when national data were lacking and parties did not object to the use of background data (see Slootweg *et al.*, 2005a,b).

***Figure 3.1.4***. *5th percentile critical loads of cadmium protecting against both human and environmental health effects. Most sensitive areas are shaded in red.*




Areas that are most sensitive to cadmium deposition (lower than 1 g $ha^{-1}$ $yr^{-1}$) are shaded red while green areas indicate least sensitive areas. Sensitive areas are in the east of the UK, in france, east of Germany, Belarus, central-southern Europe and east Russia. The CCE background database was not applied to Norway, Denmark and Spain upon request of these parties. The Ukraine submitted data from sites in the southern part of this country.

### 3.1.4 Exceedance of critical loads of cadmium

Accumulated Average Exceedances for Cd have been calculated for human health, ecosystem and the minimum of both endpoints.

Table 3.1.2 illustrates that areas at risk of adverse effects of cadmium in 2000 are in Macedonia, and Bulgaria. Figure 3.1.5. confirms exceedances in grid cells that are scattered in central-southern Europe and Russia (e.g. see 2020-CLE). Overall in Europe the area at risk reduces from 0.34% in 2000 to about nihil in 2020.
However, it should be noted that the deposition calculations of Cd are likely to be underestimated. Also, Slootweg t al. (2005) concluded that additional loads from manure and/or fertiliser will extend the area and magnitude of exceedance in central Europe.

***Table 3.1.2*** *Percentage of the area at risk of health or environmental effects caused by deposition of cadmium in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 10,082 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 61,371 | 0.00 | 0.00 | 0.00 | 0.00 |
| BA | 30,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 5,237 | 0.00 | 0.00 | 0.00 | 0.00 |
| BG | 48,330 | 15.55 | 2.57 | 2.35 | 0.00 |
| BY | 121,128 | 0.01 | 0.01 | 0.01 | 0.01 |
| CH | 11,611 | 0.00 | 0.00 | 0.00 | 0.00 |
| CY | 8,148 | 0.24 | 0.21 | 0.13 | 0.00 |
| CZ | 25,136 | 0.04 | 0.00 | 0.00 | 0.00 |
| DE | 290,003 | 0.03 | 0.02 | 0.02 | 0.01 |
| DK | 5,280 | 0.00 | 0.00 | 0.00 | 0.00 |
| EE | 29,398 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 99,616 | 0.00 | 0.27 | 0.27 | 0.27 |
| FI | 255,890 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 170,638 | 0.01 | 0.00 | 0.00 | 0.00 |
| GB | 50,075 | 0.00 | 0.00 | 0.00 | 0.00 |
| GR | 30,989 | 1.07 | 0.86 | 0.00 | 0.00 |
| HR | 23,666 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 10,560 | 0.06 | 0.86 | 0.86 | 0.55 |
| IE | 4,193 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 278,155 | 0.00 | 0.00 | 0.00 | 0.00 |
| LT | 18,099 | 0.00 | 0.00 | 0.00 | 0.00 |
| LU | 705 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 35,898 | 0.00 | 0.00 | 0.00 | 0.00 |
| MD | 2,227 | 0.00 | 0.00 | 0.00 | 0.00 |
| MK | 12,068 | 17.13 | 14.89 | 5.00 | 0.00 |
| NL | 22,312 | 0.00 | 0.00 | 0.00 | 0.00 |
| NO | 126,685 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 88,383 | 0.00 | 0.00 | 0.00 | 0.00 |
| PT | 14,572 | 0.00 | 0.00 | 0.00 | 0.00 |
| RO | 89,580 | 1.99 | 0.01 | 0.00 | 0.00 |
| RU | 1,818,725 | 0.09 | 0.43 | 0.10 | 0.02 |
| SE | 173,482 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 13,538 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 19,253 | 1.06 | 0.28 | 0.24 | 0.21 |
| UA | 18,007 | 0.00 | 0.00 | 0.00 | 0.00 |
| YU | 43,858 | 0.92 | 0.85 | 0.00 | 0.00 |
| EU25 | 1,710,932 | 0.04 | 0.04 | 0.03 | 0.02 |
| CLRTAP Europe | 4,067,625 | 0.34 | 0.29 | 0.10 | 0.02 |

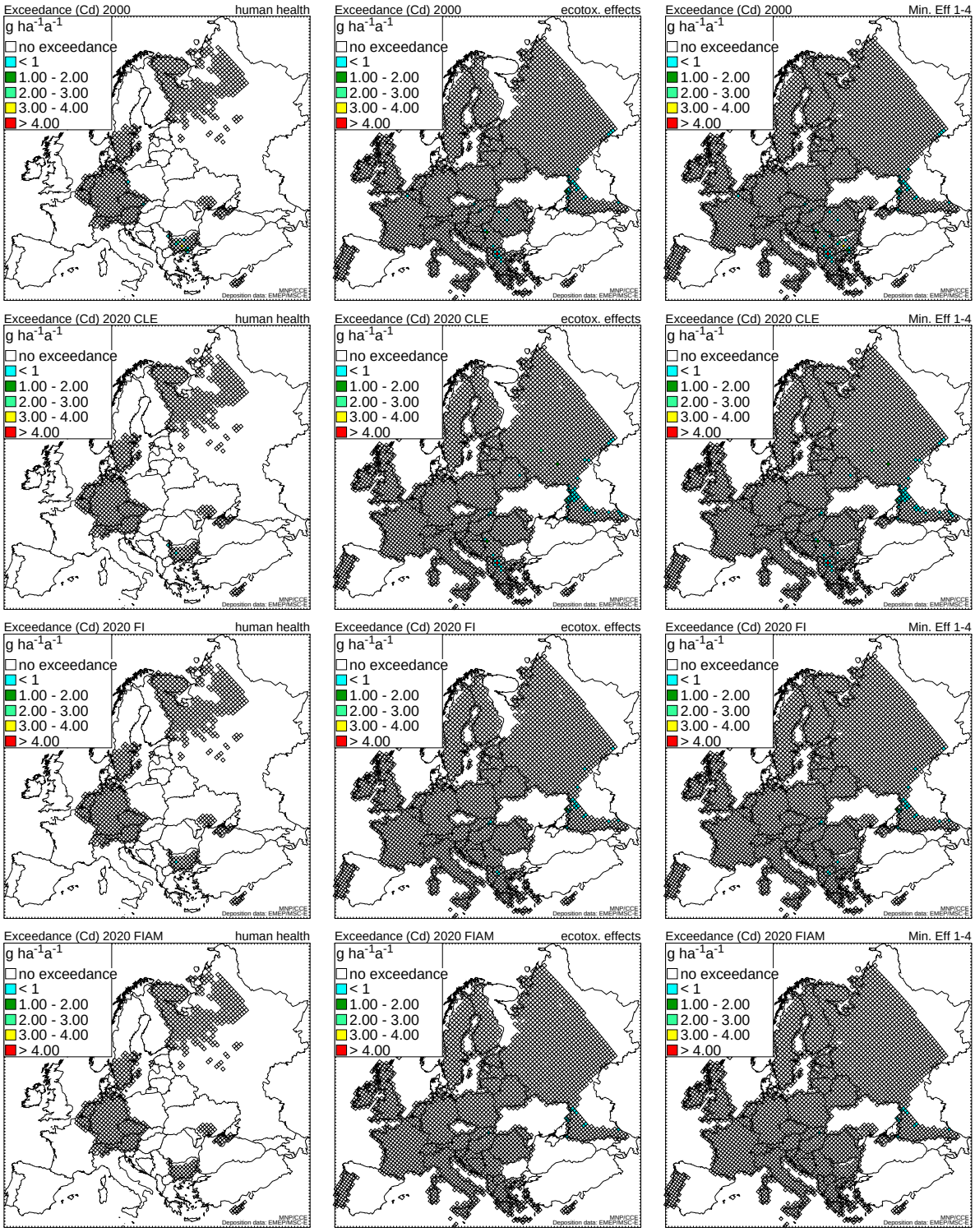



***Figure 3.1.5*** *Accumulated Average Exceedances of the critical loads of cadmium with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4).*
*The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, as agreed in the Working Group of Effects. White gridcells indicate no exceedence, whereas white areas without grids indicate no data.*

# 3.2 Mercury

## 3.2.1 Emissions

***Table 3.2.1*** *National Mercury emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 203 | 195 | 199 | 193 | 165 |
| Armenia | | 164 | 197 | 200 | 194 | 180 |
| Austria | | 1151 | 1076 | 1210 | 1210 | 805 |
| Azerbaijan | | 984 | 1174 | 1159 | 1153 | 311 |
| Belarus | | 363 | 409 | 428 | 400 | 302 |
| Belgium | | 2260 | 1407 | 1483 | 1460 | 1051 |
| Bosnia-Herzegovina | | 1982 | 1841 | 1664 | 1552 | 530 |
| Bulgaria | | 4186 | 3635 | 3587 | 3587 | 2041 |
| Croatia | | 410 | 403 | 388 | 375 | 230 |
| Cyprus | | 593 | 672 | 778 | 778 | 438 |
| Czech Republic | | 3840 | 2658 | 2357 | 2357 | 1848 |
| Denmark | | 4925 | 2477 | 1837 | 1837 | 1336 |
| Estonia | | 553 | 437 | 344 | 344 | 81 |
| Finland | | 500 | 422 | 526 | 526 | 270 |
| France | | 13375 | 11643 | 14194 | 14194 | 6571 |
| Georgia | | 253 | 305 | 314 | 305 | 239 |
| Germany | | 56014 | 40479 | 40234 | 40234 | 24486 |
| Greece | | 6650 | 7784 | 7020 | 7004 | 4133 |
| Hungary | | 4197 | 3355 | 3033 | 3033 | 1983 |
| Iceland | | 109 | 106 | 99 | 87 | 63 |
| Ireland | | 1763 | 1485 | 1412 | 1351 | 928 |
| Italy | | 10156 | 9065 | 8976 | 8931 | 6708 |
| Kazakhstan | | 17042 | 19516 | 17970 | 17051 | 8865 |
| Kyrgyzstan | | 618 | 732 | 700 | 672 | 494 |
| Latvia | | 148 | 240 | 453 | 453 | 128 |
| Lithuania | | 603 | 602 | 620 | 620 | 465 |
| Luxembourg | | 275 | 290 | 293 | 293 | 210 |
| Macedonia (Form. Yug. Rep. of) | | 1843 | 1793 | 1738 | 1664 | 1166 |
| Moldova (Rep. of) | | 146 | 142 | 131 | 131 | 50 |
| Netherlands | | 578 | 424 | 412 | 412 | 312 |
| Norway | | 996 | 998 | 1202 | 1202 | 746 |
| Poland | | 25600 | 22502 | 21361 | 21361 | 11278 |
| Portugal | | 6796 | 6146 | 6655 | 6655 | 4642 |
| Romania | | 9158 | 8535 | 9115 | 9115 | 4475 |
| Russia | | 80122 | 92713 | 91957 | 86531 | 40779 |
| Slovak Republic | | 4371 | 3410 | 3700 | 3700 | 3060 |
| Slovenia | | 644 | 571 | 584 | 584 | 312 |
| Spain | | 21788 | 18520 | 13876 | 13837 | 11193 |
| Sweden | | 746 | 779 | 1257 | 1257 | 582 |
| Switzerland | | 2630 | 2168 | 2271 | 2271 | 1962 |
| Turkey | | 18247 | 22337 | 27319 | 25524 | 18248 |
| Ukraine | | 22334 | 23477 | 22880 | 21592 | 14134 |
| United Kingdom | | 8793 | 5366 | 5350 | 5290 | 4006 |
| Yugoslavia (Fed. Rep. of) | | 5484 | 5343 | 5001 | 4659 | 1738 |
| ***Total (tonnes/yr)*** | | ***344*** | ***328*** | ***326*** | ***316*** | ***184*** |

## 3.2.2 Depositions

Depositions for Hg are calculated for the 2000 official/TNO emissions and the three emission scenario's for 2020, all including natural and historic emissions.

Concentrations and depositions of mercury were simulated on the base of official/TNO emissions. Modelled Total Gaseous Mercury (TGM) concentrations at measurement stations well agree with measured values (Figure 3.2.1.). Since TGM concentrations are mainly controlled by incoming air masses through model domain boundaries, the minor differences in TGM concentrations derived from two different emission scenarios are not surprising.

Concentrations of mercury in precipitation were overestimated by the model by about 30% (Figure 3.2.2.). On the level of individual stations the overestimation varies from 13 to about 40%, which is comparable with the uncertainly of the model. High correlation coefficient (0.97) means that mercury levels were well captured by the model, at least in the region where measurement stations are located (north of Germany, the Netherlands, Sweden, and southern Norway).

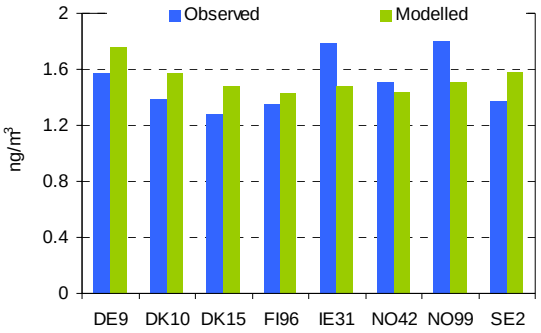



***Figure 3.2.1*** *Comparison of observed and modelled TGM concentrations*

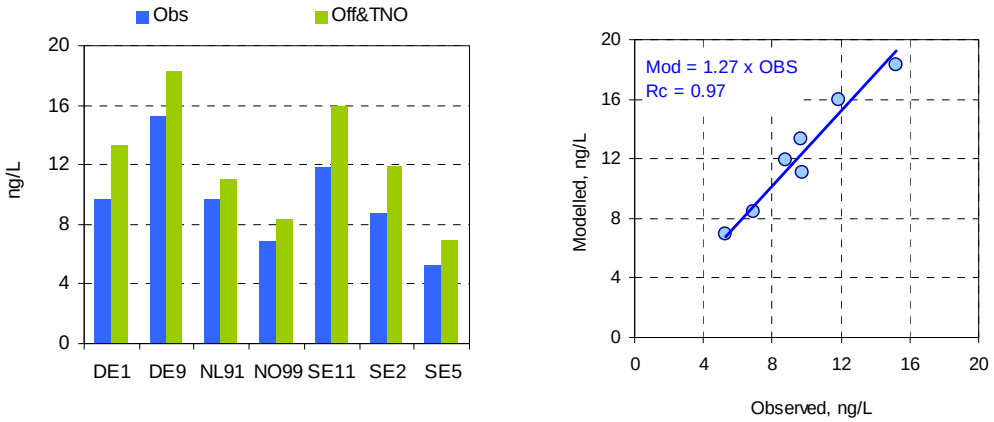



***Figure 3.2.2*** *Comparison of observed and modelled mercury concentrations in precipitation based on official/TNO emissions for 2000. Natural and historic emissions are included.. Rc – correlation coefficient*

## 3.2.3 Critical loads

Figure 3.2.3 shows that the ecosystems that are most sensitive to Hg (in grid cells shaded red) are in the north-eastern part of Europe. Relatively large areas of ecosystems with critical loads between 0.10 and 0.2 g $ha^{-1}$ $yr^{-1}$ are in the BeNeLux, France, Germany, Poland and the UK.

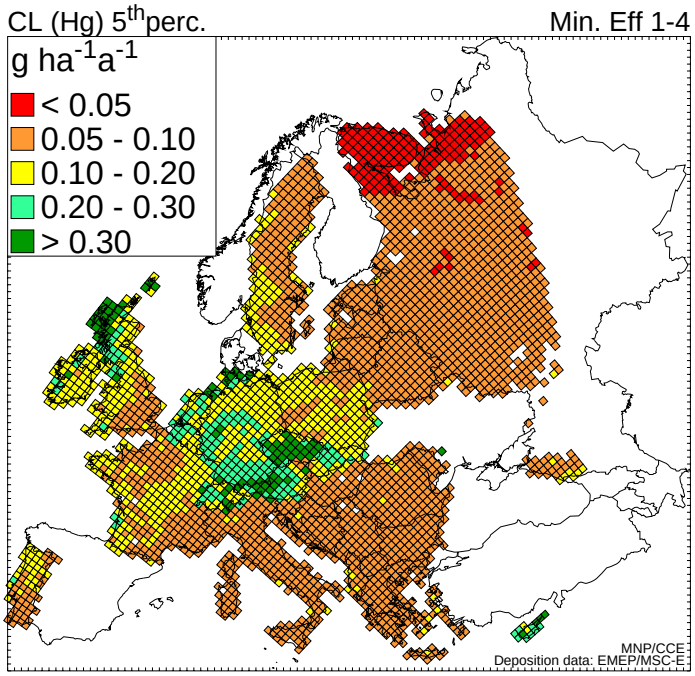



***Figure 3.2.3*** *5th percentile critical loads of mercury protecting against both human and environmental health effects. Areas that are most sensitive are shaded in red. (see Slootweg et al. 2005a,b for more details)*

## 3.2.4 Exceedance of critical loads of mercury

Accumulated Average Exceedances for Hg have been calculated for human health, ecosystem and the minimum of both endpoints.

Both Table 3.2.2 and Figure 3.2.4 indicate that the computed risk of adverse effects of mercury is both widespread and high in magnitude. More than 70% (Table 3.2.2.) of the European ecosystem area at risk of mercury deposition in 2000 and remains at risk in 2020, irrespective of the scenario.

The increase in Europe of the computed area at risk between 2000 and 2020-CLE is caused by a significant computed increase both in magnitude and distribution of mercury deposition over Russia in 2020. A possible cause is described in section 2.2.2 where the officially reported emissions in 2000 are compared to the TNO estimates in 2020.

Overall, one can state that earlier findings are confirmed that mercury contributes significantly to both the size of the European area at risk and the magnitude of the exceedances.
Finally, In addition to critical loads also critical concentrations of mercury in precipitation (see Table 2.3.1, effect 5) were computed for those countries that provided necessary data (Belgium, Finland, Sweden). These were used to calculate the Accumulated Average Concentrations (Slootweg et al. (2005a; pp 14). Results (not shown) indicate high exceedances in 2000 of which the magnitude and distribution modestly diminish in 2020 under the FIAM scenario. The increase of area at risk between 2000 and 2020 is caused by the

***Table 3.2.2*** *Percentage of the area at risk of health or environmental effects caused by deposition of mercury in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 10,082 | 98.72 | 97.51 | 97.09 | 85.02 |
| AT | 61,371 | 24.11 | 18.99 | 18.99 | 8.93 |
| BA | 30,726 | 99.56 | 99.46 | 99.46 | 97.44 |
| BE | 5,228 | 100.00 | 100.00 | 100.00 | 91.41 |
| BG | 42,512 | 100.00 | 100.00 | 100.00 | 100.00 |
| BY | 86,812 | 100.00 | 100.00 | 100.00 | 100.00 |
| CH | 11,611 | 70.97 | 68.35 | 68.35 | 57.73 |
| CY | 8,148 | 7.65 | 5.37 | 5.28 | 2.92 |
| CZ | 25,136 | 1.29 | 1.47 | 1.47 | 1.25 |
| DE | 290,003 | 34.73 | 32.80 | 32.79 | 29.62 |
| DK | 5,280 | 99.16 | 99.48 | 99.48 | 98.97 |
| EE | 29,398 | 80.42 | 85.12 | 85.12 | 74.46 |
| ES | 99,616 | 94.73 | 89.68 | 89.57 | 86.25 |
| FI | | | | | |
| FR | 123,923 | 87.25 | 86.59 | 86.59 | 80.79 |
| GB | 68,621 | 29.73 | 26.82 | 26.80 | 22.71 |
| GR | 30,989 | 100.00 | 99.92 | 99.92 | 99.51 |
| HR | 23,666 | 97.59 | 96.58 | 96.58 | 91.13 |
| HU | 10,560 | 100.00 | 100.00 | 100.00 | 100.00 |
| IE | 4,193 | 42.83 | 37.29 | 35.71 | 26.93 |
| IT | 94,729 | 98.92 | 98.70 | 98.67 | 98.12 |
| LT | 18,099 | 97.34 | 99.12 | 99.12 | 96.28 |
| LU | 705 | 100.00 | 100.00 | 100.00 | 100.00 |
| LV | 35,898 | 90.61 | 93.78 | 93.78 | 88.24 |
| MD | 2,227 | 100.00 | 100.00 | 100.00 | 100.00 |
| MK | 12,068 | 100.00 | 100.00 | 100.00 | 99.20 |
| NL | 2,842 | 100.00 | 100.00 | 100.00 | 86.19 |
| NO | 126,685 | 35.87 | 39.20 | 38.93 | 23.94 |
| PL | 88,383 | 99.99 | 100.00 | 100.00 | 99.39 |
| PT | 14,572 | 90.32 | 92.50 | 91.84 | 88.41 |
| RO | 89,580 | 99.97 | 99.97 | 99.97 | 99.89 |
| RU | 950,933 | 85.31 | 95.83 | 95.46 | 88.42 |
| SE | 152,074 | 77.75 | 77.44 | 77.43 | 61.47 |
| SI | 13,538 | 98.50 | 98.46 | 98.46 | 92.64 |
| SK | 19,253 | 89.60 | 88.60 | 88.60 | 62.22 |
| UA | 0 | 0.00 | 0.00 | 0.00 | 0.00 |
| YU | 43,858 | 99.89 | 99.87 | 99.87 | 96.39 |
| EU25 | 1,202,560 | 67.74 | 66.52 | 66.49 | 60.86 |
| CLRTAP Europe | 2,633,320 | 76.71 | 80.09 | 79.92 | 73.87 |

Figure 3.2.4 shows that areas with the highest Accumulated Average Exceedance remain in the border area of Germany and the Netherlands in Poland and south-eastern Russia in 2020 under FIAM.

The increase of the area at risk of mercury between 2000 and 2020, especially in Russia, is caused by the difference in the manner in which emissions from 2000 (officially reported) and 2020 (derived by TNO) are obtained, as described in chapter 2.2.2.

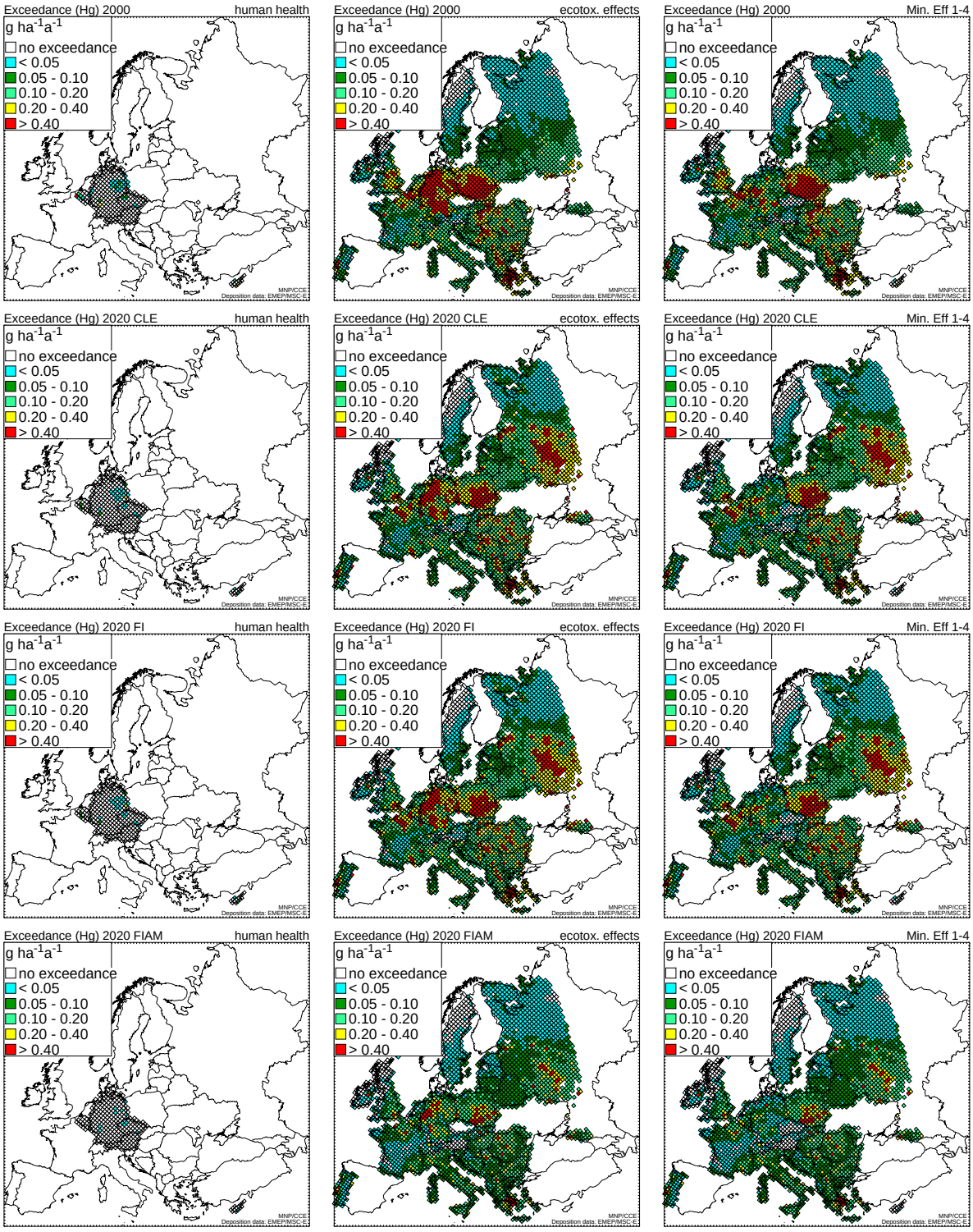



***Figure 3.2.4*** *Accumulated Average Exceedances of the critical loads of mercury with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4)*
*The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, as agreed in the Working Group of Effects. White gridcells indicate none exceedence, whereas white areas without grids indicate no data.*

# 3.3 Lead

## 3.3.1 Emissions

***Table 3.3.1***. *National Lead emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 43183 | 4660 | 6494 | 6083 | 1016 |
| Armenia | | 8274 | 1127 | 1267 | 1142 | 512 |
| Austria | | 13853 | 15098 | 14816 | 14816 | 13053 |
| Azerbaijan | | 12263 | 14902 | 16915 | 16654 | 4096 |
| Belarus | | 51039 | 53201 | 52805 | 38806 | 21202 |
| Belgium | | 133799 | 81404 | 78203 | 78203 | 46696 |
| Bosnia-Herzegovina | | 96921 | 35853 | 35449 | 15576 | 3947 |
| Bulgaria | | 214143 | 75903 | 68043 | 68043 | 22508 |
| Croatia | | 146907 | 8856 | 8684 | 8260 | 7053 |
| Cyprus | | 73973 | 5767 | 6251 | 6251 | 845 |
| Czech Republic | | 107709 | 12573 | 11169 | 11169 | 10208 |
| Denmark | | 9574 | 7983 | 5861 | 5861 | 5723 |
| Estonia | | 40730 | 16749 | 14203 | 14203 | 7311 |
| Finland | | 37500 | 36486 | 38300 | 38300 | 33447 |
| France | | 234097 | 177770 | 179014 | 179014 | 143205 |
| Georgia | | 6874 | 8414 | 10481 | 10181 | 1356 |
| Germany | | 587641 | 593980 | 647079 | 647079 | 523300 |
| Greece | | 132437 | 8905 | 8485 | 8485 | 6681 |
| Hungary | | 38659 | 32360 | 33897 | 33897 | 7081 |
| Iceland | | 197 | 196 | 173 | 172 | 153 |
| Ireland | | 8754 | 7254 | 6478 | 6478 | 6122 |
| Italy | | 908904 | 839602 | 812905 | 812905 | 105629 |
| Kazakhstan | | 600841 | 687372 | 701572 | 416719 | 231092 |
| Kyrgyzstan | | 61214 | 11989 | 12273 | 8272 | 3444 |
| Latvia | | 8230 | 791 | 1109 | 1109 | 888 |
| Lithuania | | 16121 | 21607 | 28357 | 28357 | 5290 |
| Luxembourg | | 3368 | 3884 | 4018 | 4018 | 3547 |
| Macedonia (Form. Yug. Rep. of) | | 86962 | 44958 | 45775 | 28340 | 9853 |
| Moldova (Rep. of) | | 3167 | 3063 | 3266 | 3266 | 696 |
| Netherlands | | 44070 | 39536 | 38059 | 38059 | 31547 |
| Norway | | 6035 | 6806 | 7539 | 7539 | 6455 |
| Poland | | 647499 | 282931 | 224645 | 224645 | 203291 |
| Portugal | | 39461 | 18357 | 20501 | 20501 | 18465 |
| Romania | | 604363 | 85353 | 101278 | 101278 | 22460 |
| Russia | | 5861767 | 2480810 | 2661139 | 1558887 | 545302 |
| Slovak Republic | | 74342 | 28890 | 27542 | 27542 | 21763 |
| Slovenia | | 37459 | 13141 | 12866 | 12866 | 5477 |
| Spain | | 932096 | 159389 | 163870 | 163870 | 150023 |
| Sweden | | 11811 | 11468 | 13713 | 13713 | 9510 |
| Switzerland | | 113566 | 115398 | 122507 | 122507 | 110931 |
| Turkey | | 764867 | 283553 | 382711 | 314476 | 85869 |
| Ukraine | | 1703249 | 825205 | 861659 | 500061 | 281431 |
| United Kingdom | | 192840 | 101596 | 100605 | 100605 | 63634 |
| Yugoslavia (Fed. Rep. of) | | 299771 | 51478 | 58326 | 42894 | 8484 |
| ***Total (tonnes/yr)*** | | ***15021*** | ***7317*** | ***7650*** | ***5761*** | ***2791*** |

## 3.3.2. Depositions

Depositions for Pb are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated Pb depositions are most probably an underestimation.

Modelling of lead depositions was carried out on the basis of two emission data sets for 2000: Official/TNO data and ESPREME expert estimates. Both concentrations in air and in precipitation based on Official/TNO emissions were underestimated by around 40% compared to measurements (Figure 3.3.1.). Significant correlation coefficients indicate that the spatial pattern of concentrations in air in precipitation were captured by the model. The reason of the underestimation may be connected with the underestimation of anthropogenic emission or natural and historic emissions. In the case of ESPREME emissions the underestimation is relatively small (20 – 25%). This underestimation is comparable with the estimates of model uncertainties of modelling of aerosol-bound heavy metals [Travnikov and Ilyin, 2005]. These comparisons demonstrate that ESPREME-based depositions better reproduce actual measured pollution levels, compared to the results based on Official/TNO data.

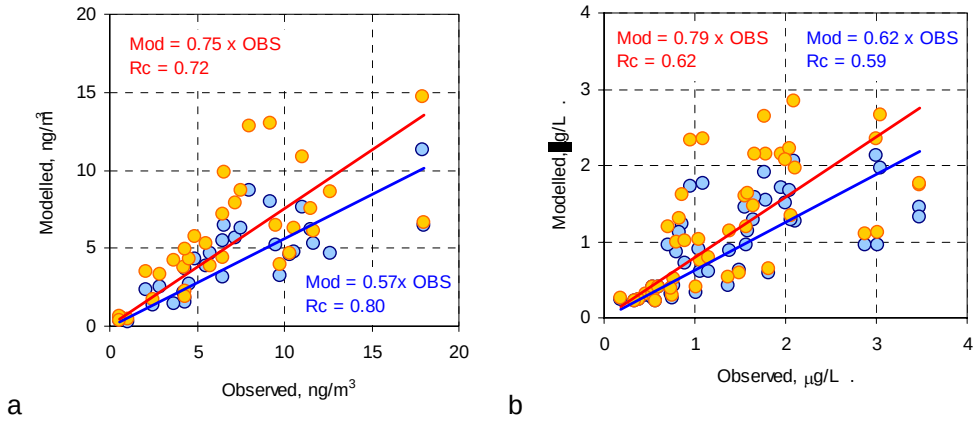



***Figure 3.3.1*** *Comparison of modelled and measured lead concentrations in air (a) and concentrations in precipitation (b), using official/ TNO (blue) and ESPREME (red) emissions. Natural and historical emissions are included. Rc = correlation coefficient*

The performance of the model is different for individual stations. An example showing the comparison of modelled concentrations of lead in air based on ESPREME emissions with measurements is presented in Figure 3.3.2 For each station contributions of anthropogenic emissions, natural and historical emissions and boundary concentrations are singled out. As can be seen, the contribution of boundary concentrations is minor at all (with few exceptions) stations. Instead, the contribution of natural emissions to air concentrations at monitoring stations is considerable, ranging from almost 20 to about 50%. The addition of natural and historic emissions turns out to significantly improve the comparison results. For example, at DE1, DE9, and Danish stations the modelled and measured concentrations became almost the same. However, at some stations (DE4, NL9) the use of anthropogenic emission alone provides better agreement with measurements. If official/TNO emissions are used, the model demonstrated better agreement with measurements at DE4, NL9 (Figure 3.3.3.). For some stations, e.g. those located in Czech Republic, Slovakia, Latvia, the use of natural emissions improves the comparison results, but modelled concentrations still remains well below the

observed values. Possibly, inadequate spatial allocation of emissions over country's area could contribute to discrepancies between measurements and model results. Further activity should be aimed at a more detailed analysis of the comparison results based on different emission data sets.

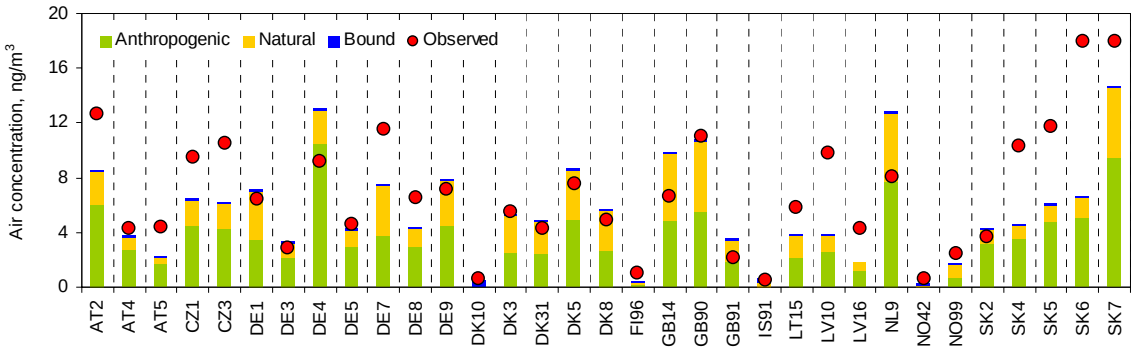



***Figure 3.3.2*** *Contribution of anthropogenic (ESPREME) emissions, natural emission and boundary concentrations to modelled annual mean air concentrations of lead*

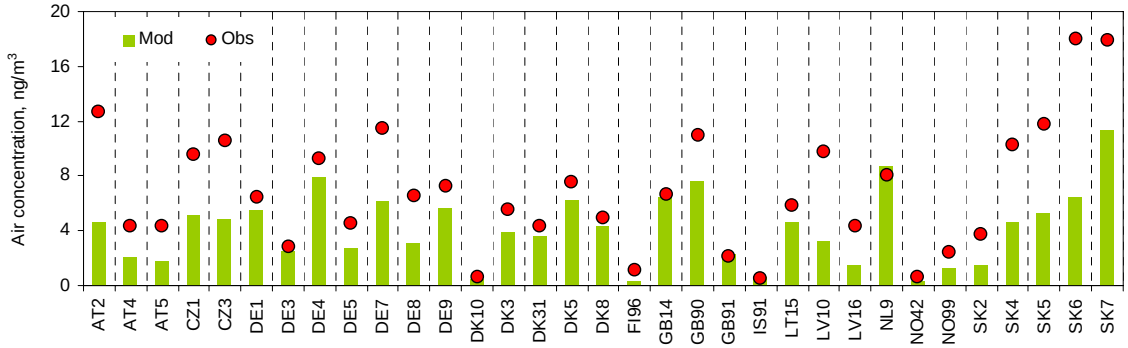



***Figure 3.3.3*** *Comparison of modelled and measured concentrations of lead in air. Official/TNO emissions and natural and historical emission.*

## 3.3.3. Critical loads

Areas most sensitive to lead are in the northern and eastern parts of Europe (see red shadings in Figure 3.3.4) But scattered areas are also in the central Europe, France, Italy and the United Kingdom. Figure 3.3.4 illustrates the range of critical loads which would lead to a computed percentage of 95 % of the ecosystems in each grid cell, provided the lower bound of the interval is not exceeded by lead deposition.

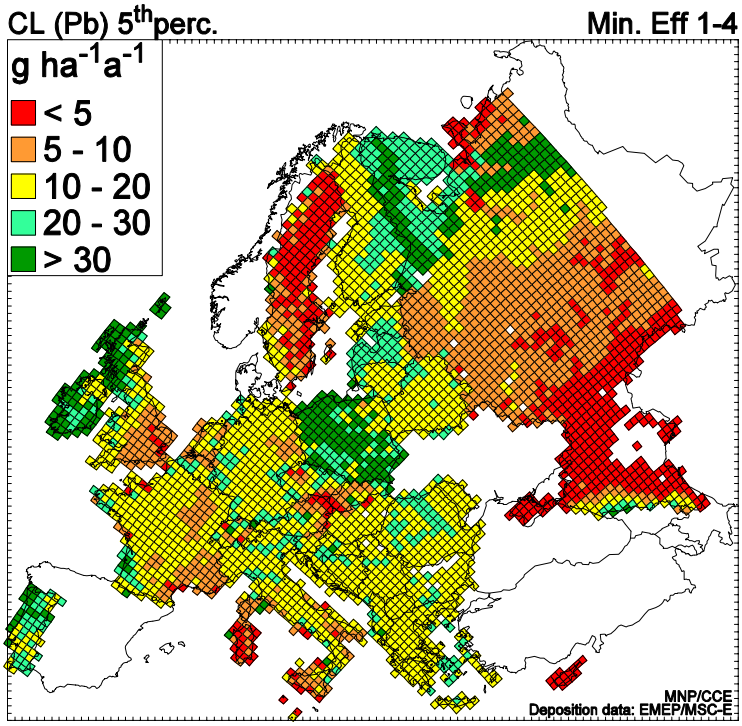



***Figure 3.3.4*** *5th percentile critical loads of lead protecting against both human and environmental health effects. Most sensitive areas are shaded in red (see Slootweg 2005a, b for further details)*

## 3.3.4 Exceedance of critical loads of lead

Accumulated Average Exceedances for Pb have been calculated for human health, ecosystem and the minimum of both endpoints.

Both Table 3.3.2 and Figure 3.3.5 indicate the significance of the area at risk on one hand and the magnitude and distribution of the exceedance on the other hand.

Comparison of the risk in 2000 of either human or environmental effects (Figure 3.3.5. top right) to 2020-FIAM (Figure 3.3.5 bottom right) shows that large parts of Europe remain subject to the risk of atmospheric deposition of lead. In 2000 about 42% of the ecosystem area is at risk compared to about 32% in 2020 under the Full Implementation scenario (see Table 3.3.2). A set of possible additional measures as indicated by TNO would reduce the risk further to 19%. However, it should be noted that the deposition calulations of Pb are likely to be underestimated. Also, Slootweg et al. (2005) concluded that additional loads from from manure and/or fertiliser will extend the area and magnitude of exceedance in Europe.

***Table 3.3.2*** *Percentage of the area at risk of health or environmental effects caused by deposition of lead in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 10,082 | 31.08 | 19.22 | 17.72 | 3.31 |
| AT | 61,371 | 25.27 | 23.00 | 22.87 | 14.79 |
| BA | 30,726 | 59.24 | 53.22 | 48.52 | 9.60 |
| BE | 5,237 | 58.96 | 58.86 | 58.86 | 57.10 |
| BG | 48,330 | 100.00 | 87.77 | 82.27 | 44.93 |
| BY | 121,128 | 18.19 | 5.67 | 1.87 | 0.07 |
| CH | 11,611 | 49.15 | 53.88 | 53.88 | 34.74 |
| CY | 8,148 | 90.00 | 76.78 | 75.47 | 57.12 |
| CZ | 25,136 | 64.73 | 59.23 | 59.12 | 38.31 |
| DE | 290,003 | 54.82 | 53.17 | 53.12 | 46.23 |
| DK | 5,280 | 27.63 | 25.48 | 24.93 | 20.06 |
| EE | 29,398 | 4.95 | 2.80 | 1.03 | 0.03 |
| ES | 99,616 | 58.07 | 57.24 | 57.24 | 19.86 |
| FI | 255,890 | 0.34 | 0.47 | 0.29 | 0.14 |
| FR | 170,638 | 71.67 | 69.49 | 69.48 | 60.66 |
| GB | 50,075 | 17.80 | 16.98 | 16.98 | 16.17 |
| GR | 30,989 | 92.28 | 64.56 | 61.00 | 18.53 |
| HR | 23,666 | 67.03 | 65.40 | 65.07 | 18.41 |
| HU | 10,560 | 89.47 | 80.09 | 78.08 | 23.66 |
| IE | 4,193 | 1.79 | 0.00 | 0.00 | 0.00 |
| IT | 278,155 | 62.04 | 60.28 | 60.01 | 34.34 |
| LT | 18,099 | 12.56 | 5.64 | 4.32 | 0.35 |
| LU | 705 | 7.53 | 13.10 | 13.10 | 9.94 |
| LV | 35,898 | 0.40 | 0.08 | 0.02 | 0.00 |
| MD | 2,227 | 98.27 | 55.79 | 18.58 | 0.00 |
| MK | 12,068 | 82.34 | 48.53 | 37.72 | 4.16 |
| NL | 22,313 | 58.55 | 54.98 | 54.98 | 45.04 |
| NO | 126,685 | 0.00 | 0.16 | 0.00 | 0.00 |
| PL | 88,383 | 28.41 | 15.69 | 15.55 | 9.43 |
| PT | 14,572 | 53.06 | 9.18 | 9.18 | 5.47 |
| RO | 89,580 | 64.25 | 9.01 | 6.93 | 1.14 |
| RU | 1,844,700 | 41.06 | 38.24 | 30.29 | 14.56 |
| SE | 151,432 | 27.59 | 23.74 | 22.04 | 16.52 |
| SI | 13,538 | 53.44 | 49.33 | 49.33 | 15.43 |
| SK | 19,253 | 36.76 | 29.99 | 29.79 | 25.55 |
| UA | 18,002 | 98.76 | 98.76 | 98.76 | 98.76 |
| YU | 43,858 | 68.12 | 26.70 | 20.33 | 4.73 |
| EU25 | 1,688,883 | 41.99 | 38.68 | 38.29 | 26.55 |
| CLRTAP Europe | 4,071,546 | 41.69 | 36.66 | 32.51 | 18.95 |

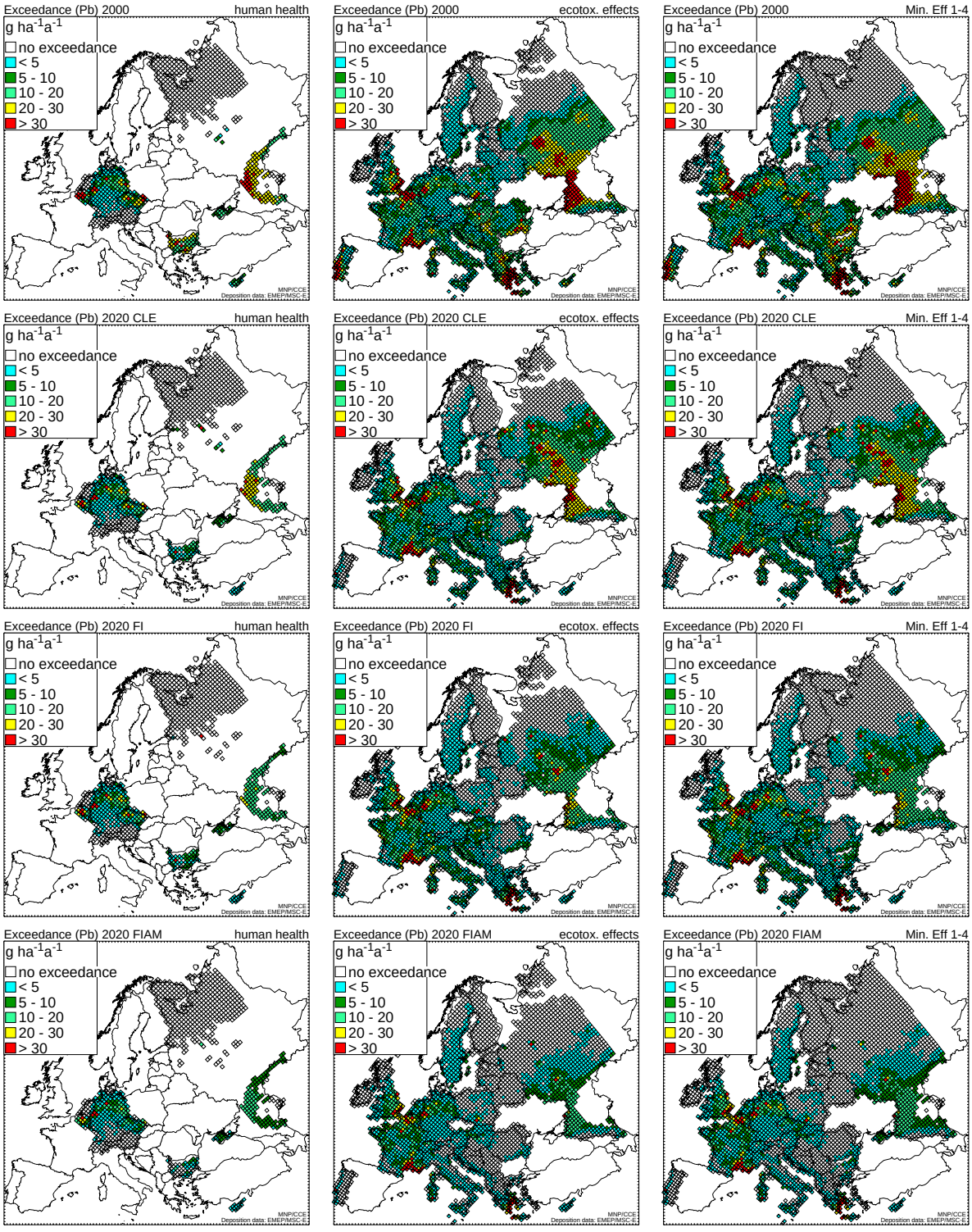



***Figure 3.3.5*** *Accumulated Average Exceedances of the critical loads of lead with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4)*
*The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, as agreed in the Working Group of Effects. White gridcells indicate none exceedence, whereas white areas without grids indicate no data.*

# 3.4 Chromium

## 3.4.1 Emissions

***Table 3.4.1.*** *National chromium emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| Country | scenario | | CLE | | FI | FIAM |
|---|---|---|---|---|---|---|
| | year | 2000 | 2010 | 2020 | 2020 | 2020 |
| Albania | | 560 | 537 | 547 | 493 | 431 |
| Armenia | | 371 | 446 | 416 | 403 | 395 |
| Austria | | 11331 | 9697 | 9741 | 9741 | 6732 |
| Azerbaijan | | 5697 | 6873 | 6734 | 6718 | 847 |
| Belarus | | 6308 | 6630 | 6216 | 3832 | 1466 |
| Belgium | | 24950 | 11004 | 9554 | 9554 | 7343 |
| Bosnia-Herzegovina | | 1749 | 1376 | 1137 | 509 | 367 |
| Bulgaria | | 7045 | 3631 | 2938 | 2938 | 1185 |
| Croatia | | 4312 | 3380 | 2894 | 2525 | 1131 |
| Cyprus | | 1580 | 1534 | 1829 | 1829 | 223 |
| Czech Republic | | 16633 | 5483 | 4306 | 4306 | 3685 |
| Denmark | | 4340 | 3757 | 3311 | 3311 | 3273 |
| Estonia | | 9686 | 3163 | 1786 | 1786 | 929 |
| Finland | | 28000 | 13537 | 15305 | 15305 | 5024 |
| France | | 259392 | 157436 | 160292 | 160292 | 90987 |
| Georgia | | 563 | 700 | 699 | 676 | 374 |
| Germany | | 73574 | 63087 | 61685 | 61685 | 54144 |
| Greece | | 34974 | 32610 | 32371 | 14814 | 4395 |
| Hungary | | 6661 | 3211 | 2879 | 2879 | 2267 |
| Iceland | | 246 | 219 | 188 | 176 | 131 |
| Ireland | | 3884 | 1947 | 1478 | 1478 | 1082 |
| Italy | | 46207 | 24612 | 19331 | 19331 | 11295 |
| Kazakhstan | | 32385 | 36015 | 31487 | 22918 | 19428 |
| Kyrgyzstan | | 854 | 1067 | 958 | 844 | 831 |
| Latvia | | 5731 | 5019 | 4849 | 4849 | 2739 |
| Lithuania | | 2405 | 2633 | 2041 | 2041 | 1826 |
| Luxembourg | | 342 | 244 | 247 | 247 | 204 |
| Macedonia (Form. Yug. Rep. of) | | 1279 | 1123 | 979 | 473 | 236 |
| Moldova (Rep. of) | | 626 | 538 | 489 | 489 | 293 |
| Netherlands | | 5558 | 5536 | 5848 | 5848 | 5150 |
| Norway | | 8814 | 4270 | 5405 | 5405 | 2510 |
| Poland | | 84300 | 40001 | 31857 | 31115 | 27195 |
| Portugal | | 12371 | 7120 | 7890 | 7890 | 5261 |
| Romania | | 11758 | 6029 | 5495 | 5495 | 1659 |
| Russia | | 1400582 | 1665307 | 2001065 | 360816 | 75876 |
| Slovak Republic | | 8059 | 3117 | 2482 | 2482 | 2054 |
| Slovenia | | 1387 | 659 | 649 | 649 | 438 |
| Spain | | 35929 | 22093 | 15315 | 15315 | 11139 |
| Sweden | | 6462 | 5763 | 4891 | 4891 | 3010 |
| Switzerland | | 3517 | 3751 | 3702 | 3702 | 3465 |
| Turkey | | 40463 | 40278 | 43124 | 25499 | 16893 |
| Ukraine | | 64528 | 68551 | 72017 | 48477 | 33193 |
| United Kingdom | | 69354 | 49416 | 54851 | 24574 | 8413 |
| Yugoslavia (Fed. Rep. of) | | 4824 | 4314 | 3836 | 1735 | 1080 |
| ***Total (tonnes/yr)*** | | ***2350*** | ***2328*** | ***2645*** | ***900*** | ***421*** |

## 3.4.2 Deposition

Depositions for Cr are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated Cr depositions are most probably an underestimation.

Observed concentrations of chromium in air and in precipitation in 2000 are underestimated by the model if official/TNO emissions are used (Fig. 3.4.1). The underestimation made up about 2.5 times for concentrations in air and about two times for concentrations in precipitation. In case of ESPREME emissions, the observed concentrations in air and in precipitation were well reproduced: regression coefficients were 0.93 and 1.06, and correlation coefficients 0.83 and 0.63, respectively. Total European ESPREME emission of chromium is about 1.5 times higher than that of official/TNO. However, the emissions in Russia, which influence on stations concentrations is relatively small, according to TNO is about 1000 t/y, while ESPREME estimate is about 400 t/y (see Fig.2.2.6.). Therefore, the ratio of total ESPREME to official/TNO emissions, excluding Russia is 3 times. The result of this large difference in emissions is that the modelled concentrations, modelled with ESPREME emissions, are higher and better fitting measurements.

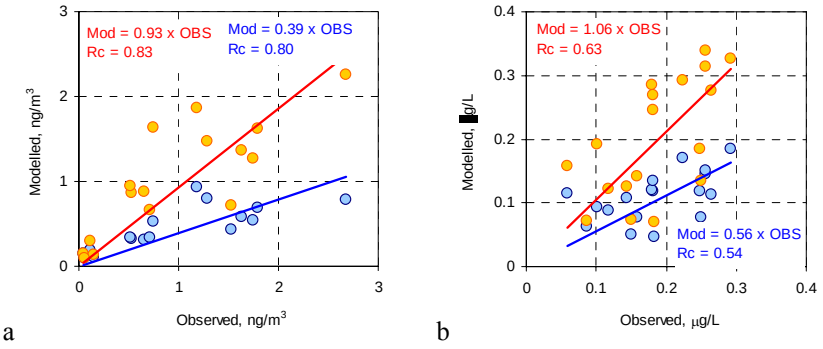



***Fig. 3.4.1*** *Comparison of modelled and measured chromium concentrations in air (a) and concentrations in precipitation (b), using official/ TNO (blue) and ESPREME (red) emissions. Natural and historical emissions are included. Rc = correlation coefficient*

## 3.4.3 Critical loads

Figure 3.4.2 provides the regional pattern in the critical load for chromium for forest systems related to eco-toxicological effects. These were computed using a fixed critical concentration of 44 µg $l^{-1}$. Because this critical concentration is almost identical to the drinking water standard of 50 $mg.m^{-3}$, critical loads and the patterns therein are very similar for the two effects. Critical loads for forests range from about 20 $g.ha^{-1}.yr^{-1}$ in dry areas to about 150 $g.ha^{-1}.yr^{-1}$ in areas with high leaching. Patterns of critical loads follow the patterns of leaching, as uptake contributes less than 2.5 % to the total critical load.

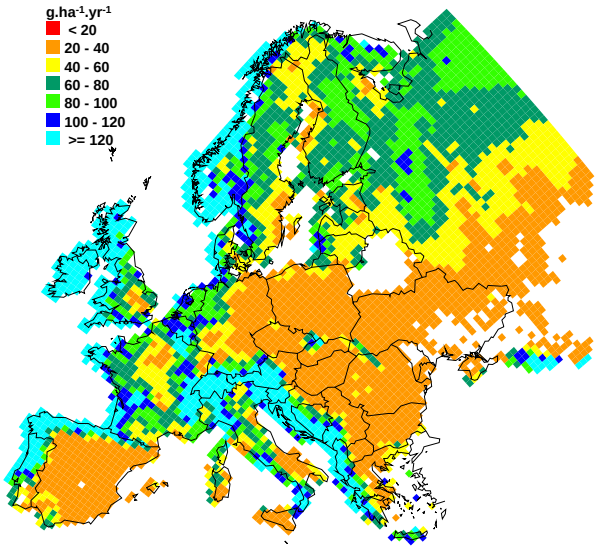



***Figure 3.4.2**. Critical loads for chromium for forests related to ecosystem functioning*

## 3.4.4 Exceedance of critical loads of chromium

Accumulated Average Exceedances for Cr have been calculated for human health, ecosystem and the minimum of both endpoints.

Table 3.4.2 shows that the percentage of the area that is computed to be at risk of chromium is limited in Europe ranging from 0.07% in 2000 to 0.01% in 2020 when the protocol is fully implemented. In the EU25 the percentages are 0.02% of an ecosystem area of 4,921,598 $km^2$.

Figure 3.4.3 shows that an Accumulated Average Exceedance of more than 4 g $ha^{-1}$ $yr^{-1}$ occurs in a few grid cells in Russia both in 2000 and in 2020 under the CLE scenario.

It can be concluded that the relative risk of impacts - i.e. the risk in 2020 compared to 2000 – is estimated to be small. However, it should be noted that the deposition calculations of Cr are likely to be underestimated.

***Table 3.4.2**: Percentage of the area at risk of health or environmental effects caused by deposition of chromium in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively*

| Country | Area ($km^2$) | 2000 (%) | 2020 CL (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 17,459 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 112,715 | 0.00 | 0.00 | 0.00 | 0.00 |
| BA | 42,948 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 33,690 | 0.00 | 0.00 | 0.00 | 0.00 |
| BG | 170,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| CH | 21,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| CZ | 126,757 | 0.00 | 0.00 | 0.00 | 0.00 |
| DE | 516,512 | 0.00 | 0.00 | 0.00 | 0.00 |
| DK | 52,987 | 0.02 | 0.02 | 0.02 | 0.01 |
| EE | 57,134 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 734,064 | 0.11 | 0.11 | 0.11 | 0.10 |
| FI | 389,983 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 808,993 | 0.00 | 0.00 | 0.00 | 0.00 |
| GB | 126,106 | 0.00 | 0.00 | 0.00 | 0.00 |
| GR | 118,953 | 0.23 | 0.26 | 0.24 | 0.22 |
| HR | 25,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 150,082 | 0.00 | 0.00 | 0.00 | 0.00 |
| IE | 16,978 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 441,533 | 0.00 | 0.00 | 0.00 | 0.00 |
| LT | 99,071 | 0.00 | 0.00 | 0.00 | 0.00 |
| LU | 4,111 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 75,240 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 33,171 | 0.00 | 0.00 | 0.00 | 0.00 |
| NO | 127,361 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 429,639 | 0.00 | 0.00 | 0.00 | 0.00 |
| PT | 103,526 | 0.00 | 0.00 | 0.00 | 0.00 |
| RO | 383,891 | 0.00 | 0.00 | 0.00 | 0.00 |
| RU | 2,065,805 | 0.21 | 0.27 | 0.00 | 0.00 |
| SE | 400,937 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 34,201 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 71,547 | 0.00 | 0.00 | 0.00 | 0.00 |
| UA | 155,570 | 0.00 | 0.01 | 0.00 | 0.00 |
| YU | 59,711 | 0.00 | 0.00 | 0.00 | 0.00 |
| EU25 | 4,921,598 | 0.02 | 0.02 | 0.02 | 0.02 |
| CLRTAP Europe | 8,007,488 | 0.07 | 0.08 | 0.01 | 0.01 |

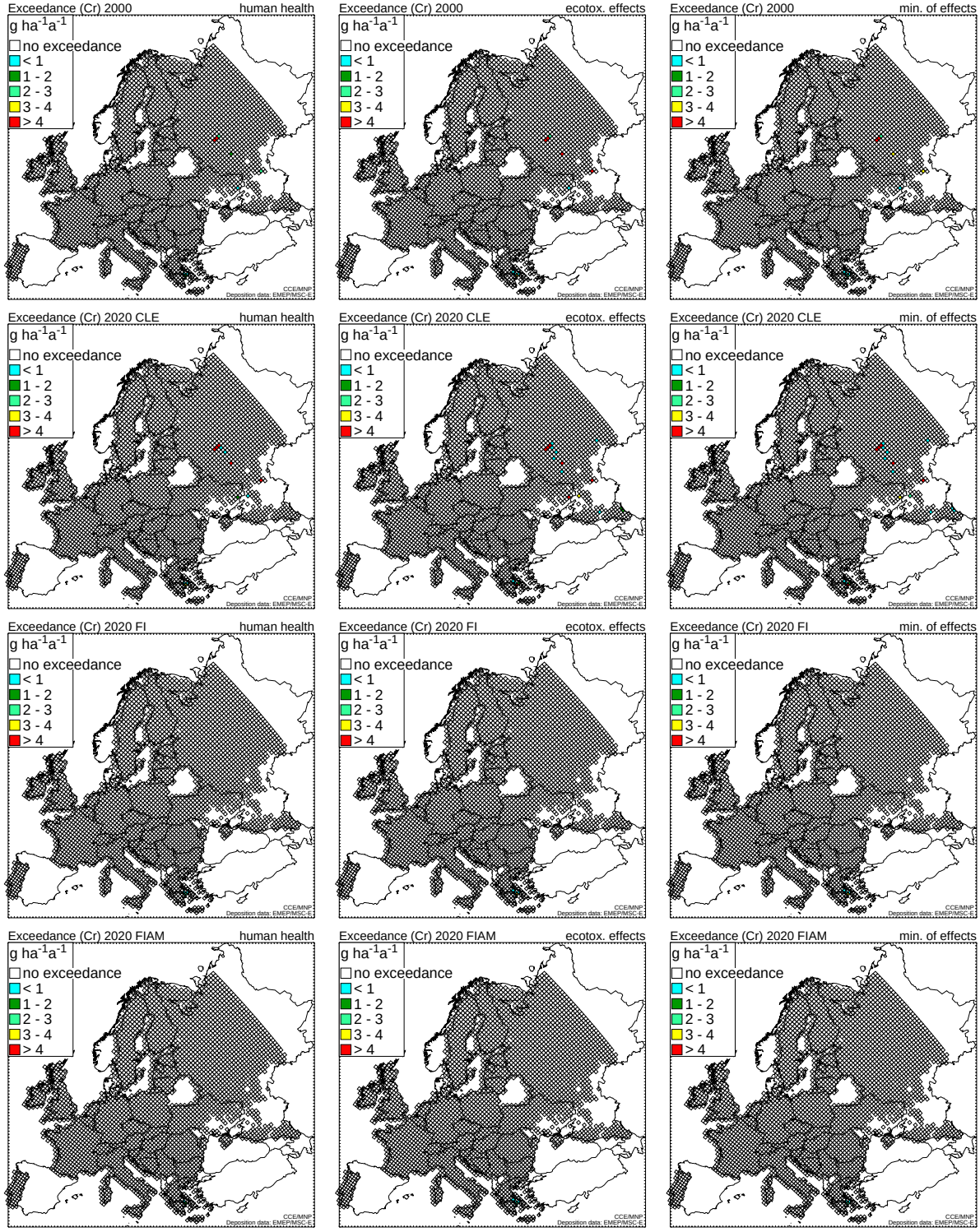



***Figure 3.4.3*** *Accumulated Average Exceedances of the critical loads of chromium with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4)*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

# 3.5 Nickel

## 3.5.1 Emissions

***Table 3.5.1*** *National nickel emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 5835 | 5629 | 5564 | 5476 | 4322 |
| Armenia | | 4127 | 4827 | 4183 | 4160 | 4153 |
| Austria | | 43058 | 32348 | 25833 | 25833 | 13186 |
| Azerbaijan | | 102477 | 123060 | 119743 | 119717 | 6780 |
| Belarus | | 94446 | 107642 | 99712 | 99190 | 11902 |
| Belgium | | 52690 | 29049 | 23849 | 23849 | 8002 |
| Bosnia-Herzegovina | | 7070 | 6102 | 5813 | 4651 | 2645 |
| Bulgaria | | 26420 | 21967 | 13126 | 13126 | 5520 |
| Croatia | | 26550 | 18998 | 11417 | 10995 | 2851 |
| Cyprus | | 25176 | 26625 | 31251 | 31251 | 332 |
| Czech Republic | | 47151 | 23597 | 14703 | 14703 | 7398 |
| Denmark | | 14246 | 7703 | 5852 | 5852 | 4935 |
| Estonia | | 7865 | 5875 | 2902 | 2902 | 856 |
| Finland | | 33200 | 16630 | 16711 | 16711 | 6877 |
| France | | 221679 | 193578 | 158743 | 158743 | 61855 |
| Georgia | | 6346 | 7730 | 7489 | 7447 | 1656 |
| Germany | | 247481 | 146987 | 114784 | 114784 | 74407 |
| Greece | | 99577 | 83849 | 74140 | 66532 | 21085 |
| Hungary | | 37241 | 20293 | 18702 | 18702 | 14061 |
| Iceland | | 3495 | 3588 | 3757 | 3747 | 3544 |
| Ireland | | 46246 | 22663 | 15380 | 15380 | 7905 |
| Italy | | 107911 | 56405 | 38763 | 38763 | 24801 |
| Kazakhstan | | 91478 | 104349 | 91925 | 74780 | 33557 |
| Kyrgyzstan | | 3435 | 5137 | 4513 | 4277 | 4247 |
| Latvia | | 11006 | 9808 | 8935 | 8935 | 860 |
| Lithuania | | 26586 | 27306 | 14433 | 14433 | 11522 |
| Luxembourg | | 680 | 468 | 390 | 390 | 385 |
| Macedonia (Form. Yug. Rep. of) | | 9511 | 6819 | 4219 | 3635 | 929 |
| Moldova (Rep. of) | | 5859 | 5734 | 5488 | 5488 | 2104 |
| Netherlands | | 53161 | 48957 | 49462 | 49462 | 35450 |
| Norway | | 56660 | 32796 | 36564 | 36564 | 19936 |
| Poland | | 251600 | 167183 | 86659 | 86464 | 61339 |
| Portugal | | 93698 | 68014 | 63073 | 63073 | 23532 |
| Romania | | 82092 | 63398 | 56505 | 56505 | 8142 |
| Russia | | 1368371 | 1627985 | 1726914 | 1019824 | 229055 |
| Slovak Republic | | 23572 | 13555 | 10698 | 10698 | 9467 |
| Slovenia | | 4352 | 3781 | 3543 | 3543 | 1514 |
| Spain | | 256487 | 147420 | 24468 | 24468 | 23690 |
| Sweden | | 17715 | 17276 | 11335 | 11335 | 4430 |
| Switzerland | | 7388 | 6925 | 6789 | 6789 | 3144 |
| Turkey | | 227099 | 185525 | 181986 | 168846 | 96207 |
| Ukraine | | 147555 | 143790 | 142213 | 105073 | 74090 |
| United Kingdom | | 125288 | 81794 | 70507 | 54241 | 25895 |
| Yugoslavia (Fed. Rep. of) | | 19819 | 16492 | 13098 | 10380 | 5749 |
| ***Total (tonnes/yr)*** | | ***4144*** | ***3750*** | ***3426*** | ***2622*** | ***964*** |

## 3.5.2. Depositions

Depositions for Ni are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated Ni depositions are likely to be underestimated.

Concentrations of nickel in air and in precipitation, based on official/TNO emissions, were underestimated by ~30% (Fig 3.5.1; Fig. 3.5.2.). This under prediction is compared to the uncertainty of the model for particulate species. Correlation coefficient for concentrations in air is high (0.87). Concentrations in air and in precipitation based on ESPREME data are somewhat (~25%) overestimated compared to measurements. Therefore, the "real" levels of pollution probably lay in between these two different model results. Correlation coefficients for concentrations in precipitation, however, are quite low in both cases. Similar to arsenic, this may require revision of wet scavenging parameters for nickel or more detailed analysis of quality of monitoring data.

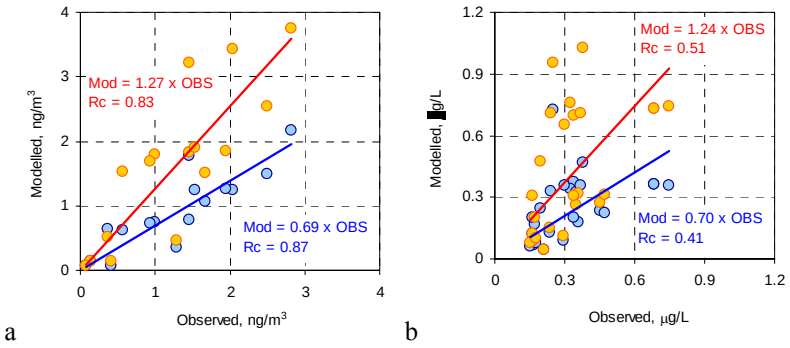



***Figure 3.5.1*** *Comparison of modelled and measured nickel concentrations in air (a) and concentrations in precipitation (b), using official/ TNO (blue) and ESPREME (red) emissions. Natural and historical emissions are included. Rc = correlation coefficient*

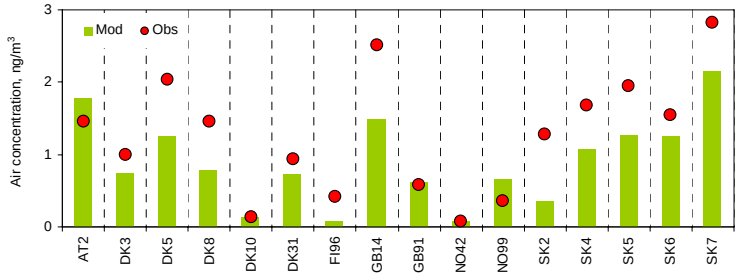



***Figure 3.5.2.*** *Comparison of modelled and measured concentrations of nickel in air. Modelling results are based on official/TNO emissions including natural and historical emission.*

## 3.5.3 Critical loads

Figure 3.5.3 provides the regional pattern in the critical load for nickel for agricultural systems related to human health effects.

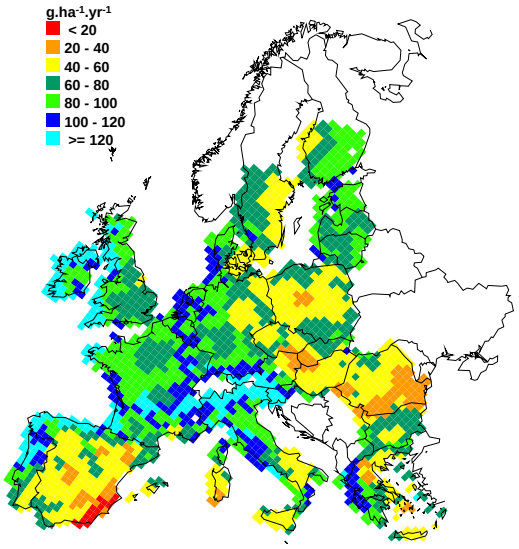



***Figure 3.5.3*** *Critical load for Ni for agriculture related to human health effects*

Critical loads related to drinking water protection are generally somewhat lower than the critical loads for ecosystem protection, because the drinking water standard of 20 $mg.l^{-1}$ is mostly lower than the critical concentration for eco-toxicological effects that varies between 25 and 700 $mg.l^{-1}$ (with a median value of about 80 $mg.l^{-1}$) for forests depending on pH and DOC concentration. Lowest critical loads are confined to regions with a low precipitation surplus in southern and eastern Europe.

## 3.5.4. Exceedance of the critical loads of nickel

Accumulated Average Exceedances for Ni have been calculated for human health, ecosystem and the minimum of both endpoints.

Table 3.5.2 shows that the percentage of the European ecosystem area that is computed to be at risk of nickel ranges from 0.71% in 2000 to 0.21% in 2020 when the protocol is fully implemented. Adding the overestimated effect of additional measures would still leave 0.03% of the area at risk. In the EU25 the percentages are 0.72% and 0.04% respectively of the ecosystem area.

Looking at the absolute magnitude of exceedances rather than area percentages shows in Figure 3.5.4 (top left map) that the risk to human health in 2000 covers a larger area in central Europe than the area at risk of ecosystem effects (to middle map). In the east of Europe and in Greece the computed risk of ecosystem effects seem to be higher than the risk to human health. The combination of the risks is shown in the top right map. Going from the top three maps (exceedances in 2000) to the bottom three maps (exceedances in 2020 under FI plus additional measures) we see that exceedances may remain in Greece.

It can be concluded that the relative risk of impacts - i.e. the risk in 2020 compared to 2000 – is not widespread and effectively reduced in 2020. However, it should be noted that deposition calculations of Ni are likely to be underestimated.

***Table 3.5.2*** *Percentage of the area at risk of health or environmental effects caused by deposition of nickel in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively.*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 17,459 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 112,715 | 0.07 | 0.00 | 0.00 | 0.00 |
| BA | 42,948 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 33,690 | 0.00 | 0.00 | 0.00 | 0.00 |
| BG | 170,043 | 0.17 | 0.01 | 0.00 | 0.00 |
| CH | 21,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| CZ | 126,757 | 0.28 | 0.00 | 0.00 | 0.00 |
| DE | 516,512 | 0.63 | 0.01 | 0.01 | 0.00 |
| DK | 52,987 | 0.12 | 0.10 | 0.10 | 0.08 |
| EE | 57,134 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 734,064 | 0.79 | 0.10 | 0.10 | 0.09 |
| FI | 389,983 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 808,993 | 0.20 | 0.07 | 0.07 | 0.01 |
| GB | 126,106 | 0.01 | 0.00 | 0.00 | 0.00 |
| GR | 118,953 | 3.22 | 2.53 | 2.42 | 1.19 |
| HR | 25,043 | 0.04 | 0.00 | 0.00 | 0.00 |
| HU | 150,082 | 0.72 | 0.00 | 0.00 | 0.00 |
| IE | 16,978 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 441,533 | 0.11 | 0.01 | 0.01 | 0.01 |
| LT | 99,071 | 0.00 | 0.00 | 0.00 | 0.00 |
| LU | 4,111 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 75,240 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 33,171 | 0.00 | 0.00 | 0.00 | 0.00 |
| NO | 127,361 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 429,639 | 4.28 | 0.03 | 0.03 | 0.00 |
| PT | 103,526 | 0.00 | 0.00 | 0.00 | 0.00 |
| RO | 383,891 | 0.51 | 0.47 | 0.47 | 0.00 |
| RU | 2,065,805 | 0.74 | 1.11 | 0.43 | 0.00 |
| SE | 400,937 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 34,201 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 71,547 | 1.07 | 0.02 | 0.02 | 0.00 |
| UA | 155,570 | 1.87 | 1.65 | 0.60 | 0.02 |
| YU | 59,711 | 0.08 | 0.00 | 0.00 | 0.00 |
| EU25 | 4,921,598 | 0.72 | 0.09 | 0.09 | 0.04 |
| CLRTAP Europe | 8,007,488 | 0.71 | 0.40 | 0.21 | 0.03 |

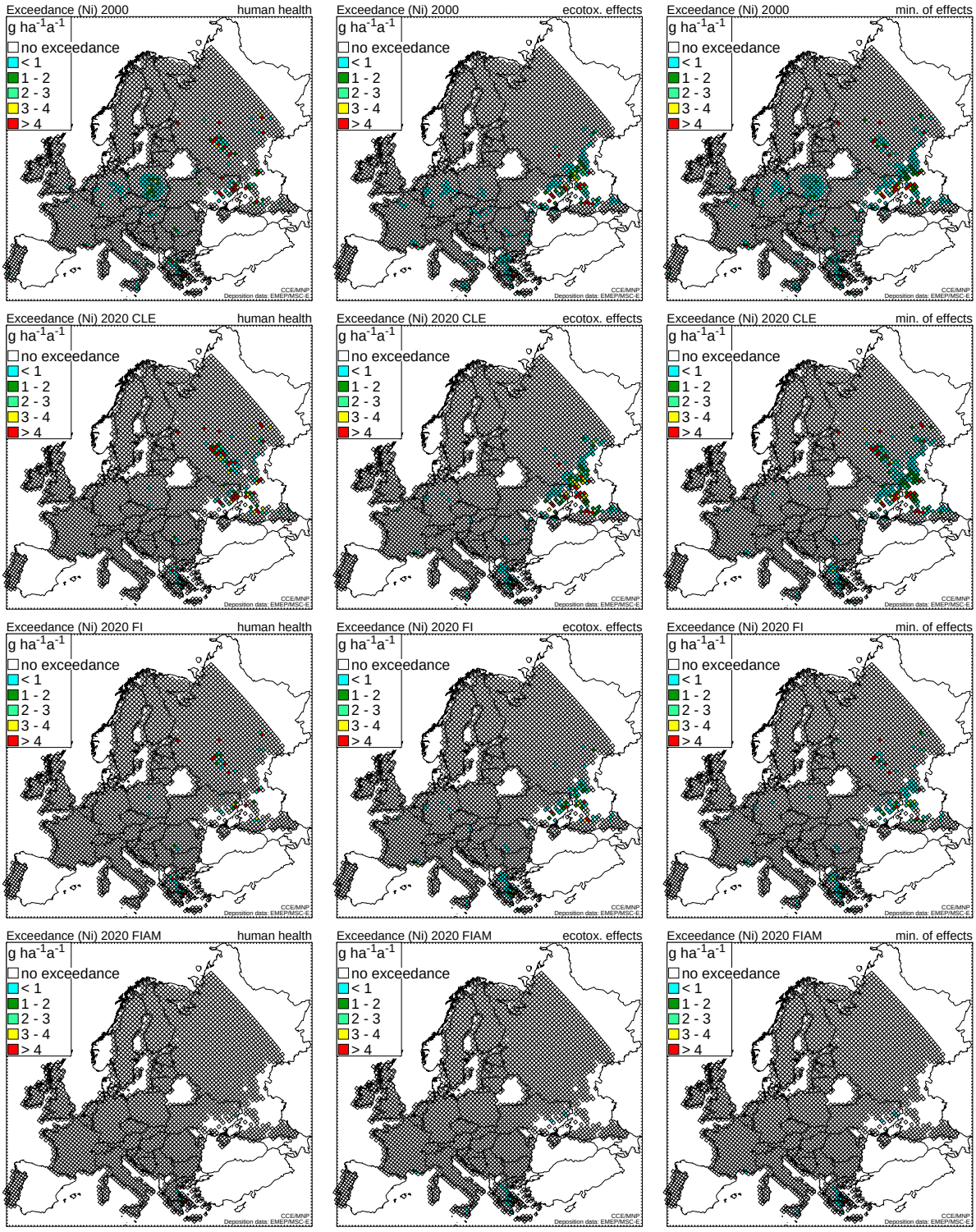



***Figure 3.5.4***. *Accumulated Average Exceedances of the critical loads of nickel with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4)*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

## 3.6 Copper

### 3.6.1 Emissions

**Table 3.6.1** *National Copper emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| Country | scenario | | CLE | | FI | FIAM |
|---|---|---|---|---|---|---|
| | year | 2000 | 2010 | 2020 | 2020 | 2020 |
| Albania | | 1176 | 1429 | 1738 | 1626 | 1559 |
| Armenia | | 553 | 663 | 686 | 685 | 683 |
| Austria | | 32595 | 37004 | 38417 | 38417 | 36503 |
| Azerbaijan | | 4767 | 5759 | 6137 | 6134 | 3853 |
| Belarus | | 14472 | 15302 | 14888 | 10155 | 8071 |
| Belgium | | 26850 | 20129 | 21132 | 21132 | 20242 |
| Bosnia-Herzegovina | | 9166 | 9025 | 8660 | 3762 | 3039 |
| Bulgaria | | 18703 | 9014 | 9474 | 9474 | 7721 |
| Croatia | | 9788 | 10265 | 11813 | 11743 | 11451 |
| Cyprus | | 1614 | 1816 | 2104 | 2104 | 1479 |
| Czech Republic | | 43703 | 22676 | 22016 | 22016 | 21577 |
| Denmark | | 11646 | 12197 | 12223 | 12223 | 12192 |
| Estonia | | 3482 | 2097 | 2216 | 2216 | 2183 |
| Finland | | 18700 | 19968 | 21975 | 21975 | 14676 |
| France | | 177165 | 187904 | 188634 | 188634 | 185490 |
| Georgia | | 2834 | 3346 | 3720 | 3692 | 3571 |
| Germany | | 273268 | 306291 | 328887 | 328887 | 296405 |
| Greece | | 18176 | 17378 | 18720 | 18720 | 17653 |
| Hungary | | 18731 | 19089 | 20890 | 20890 | 20578 |
| Iceland | | 428 | 464 | 465 | 464 | 459 |
| Ireland | | 8776 | 9728 | 10199 | 10199 | 10044 |
| Italy | | 72426 | 50898 | 48247 | 48247 | 43236 |
| Kazakhstan | | 218235 | 254541 | 281551 | 144317 | 64819 |
| Kyrgyzstan | | 2405 | 2828 | 2740 | 1941 | 1828 |
| Latvia | | 4095 | 2016 | 2616 | 2616 | 2452 |
| Lithuania | | 6745 | 9935 | 12995 | 12995 | 12853 |
| Luxembourg | | 1252 | 1483 | 1618 | 1618 | 1531 |
| Macedonia (Form. Yug. Rep. of) | | 3450 | 3451 | 3386 | 2182 | 1568 |
| Moldova (Rep. of) | | 1573 | 1603 | 1569 | 1569 | 1544 |
| Netherlands | | 15537 | 15025 | 14871 | 14871 | 14299 |
| Norway | | 19329 | 21930 | 24005 | 24005 | 21056 |
| Poland | | 374500 | 141458 | 105133 | 105133 | 92060 |
| Portugal | | 21227 | 19429 | 23267 | 23267 | 22270 |
| Romania | | 25813 | 17812 | 20874 | 20874 | 17923 |
| Russia | | 800316 | 894331 | 963938 | 582690 | 398612 |
| Slovak Republic | | 23685 | 10761 | 11284 | 11284 | 10671 |
| Slovenia | | 4463 | 4045 | 4040 | 4040 | 3957 |
| Spain | | 149544 | 110434 | 113689 | 113689 | 105166 |
| Sweden | | 15116 | 16057 | 17405 | 17405 | 15869 |
| Switzerland | | 21641 | 22579 | 21674 | 21674 | 21538 |
| Turkey | | 90054 | 99339 | 120617 | 87969 | 73117 |
| Ukraine | | 198395 | 178573 | 180078 | 113189 | 86997 |
| United Kingdom | | 48183 | 20539 | 18685 | 18685 | 15381 |
| Yugoslavia (Fed. Rep. of) | | 31223 | 31835 | 32249 | 16556 | 10898 |
| ***Total (tonnes/yr)*** | | ***2846*** | ***2642*** | ***2772*** | ***2126*** | ***1719*** |

The uncertainty of the relation between sources and magnitudes of copper emissions receives continued attention. Recently Hulskotte et al. (2006) suggested that brake wear from road traffic vehicles is an important source of diffuse atmospheric (particulate) copper emissions in Europe. They concluded that current emission inventories underestimate copper emissions and that proper inclusion of brake wear as a source of Cu may increase the copper emission estimates by 20-40%. Consequently, brake wear also contributes significantly to deposition fluxes of copper to surface waters. Hulskotte et al. (2006) estimate that brake wear emissions dominate the total emission of copper in Western Europe and reported that 75% of the atmospheric copper input in the North Sea may be due to brake wear. So, although the estimated brake wear copper emission is associated with a large uncertainty, properly including this source may significantly improve our understanding of the copper cycle in the environment. However, the late availability of these very recent results, and their preliminary nature prevented their use in this report.

**References**

Hulskotte, J.H.J. ,M. Schaap, A.J.H. Visschedijk (2006a) "Brake wear from vehicles as an important source of diffuse copper pollution" paper presented at '10th International Specialized Conference on Diffuse Pollution and Sustainable Basin Management' , 18-22 September 2006, Istanbul, Turkey.

## 3.6.2 Depositions

Depositions for Cu are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated Cu depositions are most probably an underestimation.

Calculated copper concentrations in air and in precipitation are most probably significantly underpredicted by the model (Fig. 3.6.1): regression coefficients were 0.34 and 0.14, respectively. *W.A.S. Nijenhuis et al*. [2001] simulated copper transport and depositions over the North Sea, and their modelled air concentrations underestimate measurements also by a factor of 3. Possible reasons of the underestimation are similar to those for zinc.

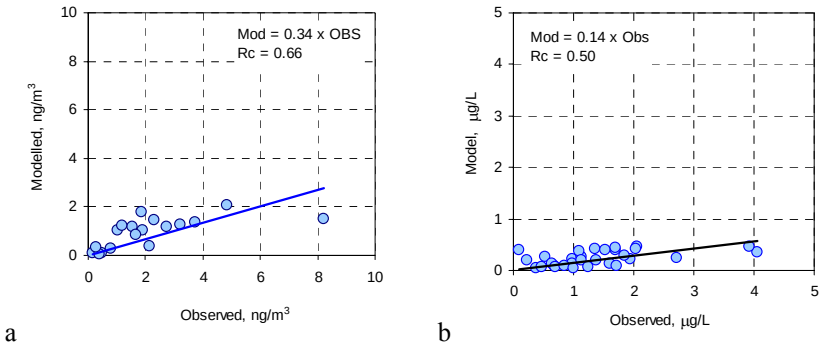



***Figure 3.6.1*** *Comparison of modelled and measured concentrations in air (a) and concentrations in precipitation (b) of copper. Modelling results are based on official/TNO emissions including natural and historical emission. Rc –correlation coefficient.*

## 3.6.3 Critical Loads

Figure 3.6.2 provides the regional pattern in the critical load for copper for forest systems related to eco-toxicological effects.

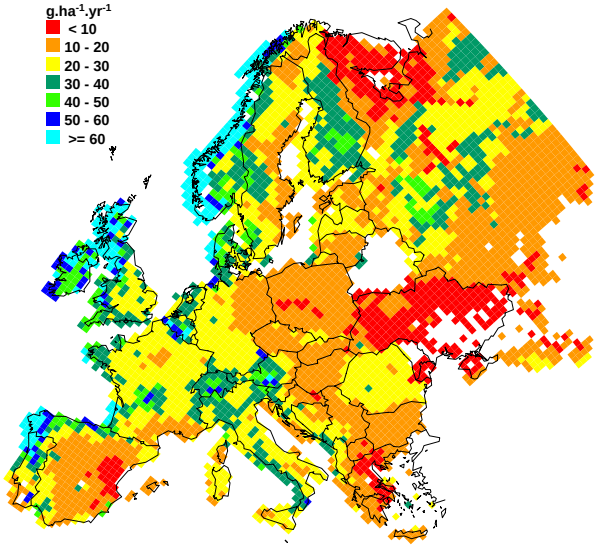



***Figure 3.6.2****. Critical load for Copper for forests related to ecosystem functioning*

Data behind Figure 3.6.2 reveals that 5 percentile critical loads for copper related to ecosystem functioning, range between 5 and 200 $g.ha^{-1}.yr^{-1}$. Highest critical loads (> 60 g $ha^{-1}$ $yr^{-1}$) are found in areas with a high precipitation surplus such as south-western Norway and Scotland, whereas low critical loads are found in areas with a very low precipitation surplus, such as central Spain. In general critical loads are higher for agriculture than for forests because of higher leaching fluxes under annual crops compared to forests (part of the year the soil is bare when growing annual crops) and because of the higher uptake of Cu by wheat that is about twice as high as the uptake by forests due to its higher yield. Because copper is a nutrient, a substantial part (up to about 50 %) of the critical load in areas with low leaching consist of uptake.

## 3.6.4. Exceedance of the critical loads of copper

Accumulated Average Exceedances for Cu have been calculated for human health, ecosystem and the minimum of both endpoints

Table 3.6.2 shows that the percentage of the European ecosystem area that is computed to be at risk of copper ranges from 0.76% in 2000 to 0.34% in 2020-FI, i.e. when the protocol is fully implemented. A relatively large area at risk (about 7%) in 2000 is in Poland. Following the FI scenario the area at risk in Poland can be reduced to about 1.5%. Please note that in France the area at risk increases from 2000 to 2020. The reason for this – already identifiable by higher 2020 deposition in comparison to 2000 – requires further exploration.

Figure 3.6.3 shows that the risk to ecosystem effects (middle column) are predominant in middle Europe. Areas where the AAE exceeds 4 g $ha^{-1}$ $yr^{-1}$ are mostly in Germany, Poland and the Ukraine. The surprising increase - from 2000 to 2020 - (noted above) of area at risk in France is reflected in the addition of two grid cells in the north of France where exceedances are between 0 and 1 g $ha^{-1}$ $yr^{-1}$

Overall it can be concluded that the relative risk of impacts – i.e. the risk in 2020 compared to 2000 – while being reduced both in area and in magnitude remains mostly in Germany, Poland and the Ukraine. However, on Europe as a whole the ecosystem at risk remains lower than 1 %. However, it should be noted that deposition computations of cupper are likely to be underestimated.

***Table 3.6.2***. *Percentage of the area at risk of health or environmental effects caused by deposition of Copper in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively.*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 17,459 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 112,715 | 0.00 | 0.00 | 0.00 | 0.00 |
| BA | 42,948 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 33,690 | 0.07 | 0.07 | 0.07 | 0.00 |
| BG | 170,043 | 1.22 | 0.19 | 0.03 | 0.01 |
| CH | 21,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| CZ | 126,757 | 0.08 | 0.02 | 0.02 | 0.00 |
| DE | 516,512 | 1.21 | 1.44 | 1.44 | 1.02 |
| DK | 52,987 | 0.00 | 0.00 | 0.00 | 0.00 |
| EE | 57,134 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 734,064 | 0.26 | 0.11 | 0.11 | 0.11 |
| FI | 389,983 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 808,993 | 0.25 | 0.34 | 0.34 | 0.33 |
| GB | 126,106 | 0.00 | 0.00 | 0.00 | 0.00 |
| GR | 118,953 | 0.11 | 0.11 | 0.10 | 0.10 |
| HR | 25,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 150,082 | 0.31 | 0.17 | 0.17 | 0.17 |
| IE | 16,978 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 441,533 | 0.00 | 0.00 | 0.00 | 0.00 |
| LT | 99,071 | 0.00 | 0.00 | 0.00 | 0.00 |
| LU | 4,111 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 75,240 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 33,171 | 0.04 | 0.00 | 0.00 | 0.00 |
| NO | 127,361 | 0.00 | 0.02 | 0.00 | 0.00 |
| PL | 429,639 | 6.99 | 1.51 | 1.49 | 0.99 |
| PT | 103,526 | 0.02 | 0.03 | 0.03 | 0.03 |
| RO | 383,891 | 0.00 | 0.00 | 0.00 | 0.00 |
| RU | 2,065,805 | 0.24 | 0.35 | 0.12 | 0.06 |
| SE | 400,937 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 34,201 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 71,547 | 2.13 | 0.70 | 0.67 | 0.62 |
| UA | 155,570 | 7.47 | 6.01 | 4.16 | 3.29 |
| YU | 59,711 | 0.22 | 0.14 | 0.00 | 0.00 |
| EU25 | 4,921,598 | 0.86 | 0.37 | 0.37 | 0.28 |
| CLRTAP Europe | 8,007,488 | 0.76 | 0.44 | 0.34 | 0.25 |

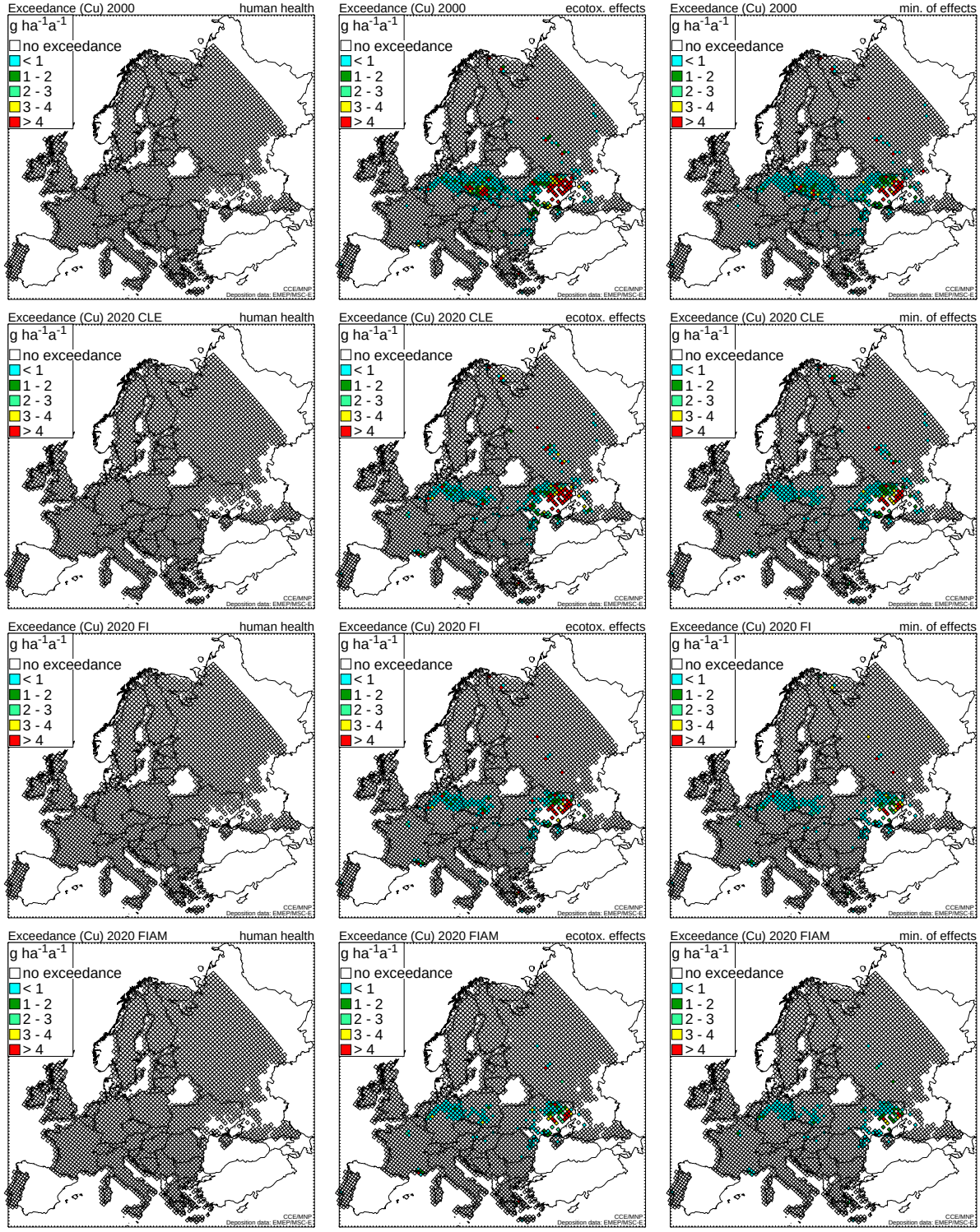



***Figure 3.6.3*** *Accumulated Average Exceedances of the critical loads of copper with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4).*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

# 3.7 Zinc

## 3.7.1 Emissions

***Table 3.7.1*** *National zinc emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| Country | scenario | | CLE | | FI | FIAM |
|---|---|---|---|---|---|---|
| | year | 2000 | 2010 | 2020 | 2020 | 2020 |
| Albania | | 4144 | 4419 | 4945 | 4232 | 4123 |
| Armenia | | 2004 | 2548 | 2679 | 2618 | 2589 |
| Austria | | 187706 | 228337 | 250095 | 250095 | 245316 |
| Azerbaijan | | 11643 | 15631 | 18951 | 18863 | 18357 |
| Belarus | | 196768 | 263051 | 257471 | 59330 | 44663 |
| Belgium | | 166330 | 112573 | 115617 | 115617 | 85918 |
| Bosnia-Herzegovina | | 68974 | 66791 | 61897 | 19496 | 12532 |
| Bulgaria | | 130185 | 44003 | 40743 | 40743 | 22798 |
| Croatia | | 61059 | 73663 | 90571 | 88800 | 87260 |
| Cyprus | | 2484 | 2051 | 2751 | 2751 | 2620 |
| Czech Republic | | 319506 | 112631 | 104580 | 104580 | 103683 |
| Denmark | | 65939 | 72094 | 67626 | 67626 | 67219 |
| Estonia | | 52963 | 16053 | 12579 | 12579 | 12549 |
| Finland | | 70600 | 72816 | 79833 | 79833 | 73037 |
| France | | 1441439 | 1430449 | 1549390 | 1549390 | 1484111 |
| Georgia | | 3720 | 4668 | 4963 | 4613 | 4540 |
| Germany | | 1657857 | 1815932 | 1982015 | 1982015 | 1900995 |
| Greece | | 76027 | 69420 | 79500 | 79500 | 76446 |
| Hungary | | 40231 | 33753 | 38688 | 38688 | 38303 |
| Iceland | | 2436 | 2485 | 2867 | 2862 | 2829 |
| Ireland | | 20476 | 26340 | 30653 | 30653 | 29674 |
| Italy | | 1430866 | 937574 | 973348 | 973348 | 704392 |
| Kazakhstan | | 1026227 | 1180709 | 1173376 | 624659 | 365133 |
| Kyrgyzstan | | 17259 | 20093 | 18525 | 10822 | 9549 |
| Latvia | | 56498 | 17459 | 18876 | 18876 | 16861 |
| Lithuania | | 63877 | 95622 | 134915 | 134915 | 134831 |
| Luxembourg | | 36697 | 43137 | 46163 | 46163 | 39576 |
| Macedonia (Form. Yug. Rep. of) | | 439638 | 439156 | 438632 | 206665 | 54196 |
| Moldova (Rep. of) | | 16515 | 8523 | 8268 | 8268 | 6957 |
| Netherlands | | 103445 | 112960 | 124380 | 124380 | 117696 |
| Norway | | 61777 | 70018 | 83343 | 83343 | 73520 |
| Poland | | 2172999 | 989804 | 794350 | 794350 | 760381 |
| Portugal | | 121127 | 86350 | 113789 | 113789 | 111384 |
| Romania | | 679514 | 134763 | 144271 | 144271 | 55815 |
| Russia | | 4831987 | 5615332 | 5984278 | 2556553 | 1459434 |
| Slovak Republic | | 59189 | 29854 | 29310 | 29310 | 28805 |
| Slovenia | | 25219 | 14203 | 15031 | 15031 | 14355 |
| Spain | | 789550 | 638620 | 700703 | 700703 | 635875 |
| Sweden | | 92328 | 122535 | 299756 | 299756 | 294449 |
| Switzerland | | 558285 | 642228 | 680738 | 680738 | 672647 |
| Turkey | | 506920 | 552363 | 617376 | 398530 | 310972 |
| Ukraine | | 1298408 | 1395731 | 1410914 | 909508 | 570528 |
| United Kingdom | | 413183 | 291599 | 287168 | 287168 | 273250 |
| Yugoslavia (Fed. Rep. of) | | 119362 | 116173 | 110173 | 49491 | 27649 |
| ***Total (tonnes/yr)*** | | ***19503*** | ***18025*** | ***19006*** | ***13766*** | ***11058*** |

## 3.7.2 Depositions

Depositions for Zn are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated Zn depositions are most probably an underestimation.

Comparison of modelled air concentrations and concentrations in precipitation for official/TNO emissions in 2000 for Zn with observations are demonstrated in Fig. 3.7.1 Both for concentrations in air and in precipitation the model underpredicts the measured parameters by an order of magnitude. Attempts to model long-range transport of zinc have been undertaken earlier by other researches [e.g., *Nijenhuis et al.*, 2001; *Alcamo et al.*, 1992; *Sofiev et al.*, 2001, *Bartnicki et al*., 1998]. The comparisons of modelled zinc concentrations and depositions with measured values, published in these papers, indicate essential underestimation of measurements. Wet depositions are underestimated by a factor 4 – 13, air concentrations – 3.5 – 10 times. The researches explain the underestimation by low available emission estimates of zinc. Besides, *J.Bartnicki et al*. [1998] assumed that quality of zinc measurements can also contribute to the underestimation. The underestimation of zinc measurements by MSCE-HM could be connected with underestimated emissions (natural or anthropogenic or both), with quality of measurements and with uncertainties of the model. These assumptions need more detailed investigation.

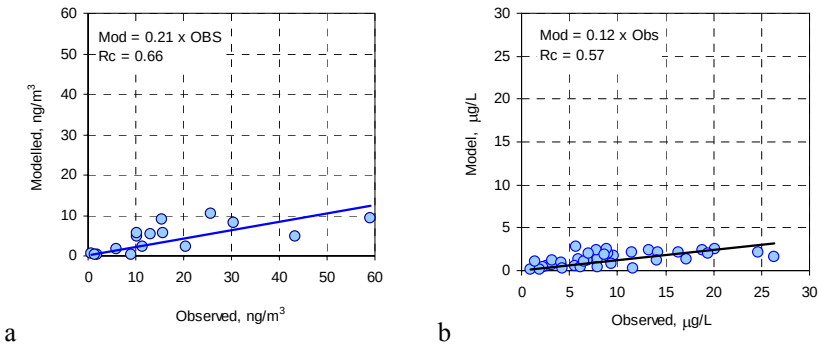



***Figure 3.7.1*** *Comparison of modelled and measured zinc concentrations: in air (a) and concentrations in precipitation (b). Modelling results are based on official/TNO emissions including natural and historical emission. Rc = correlation coefficient.*

## 3.7.3 Critical loads

Figure 3.7.2. provides the regional pattern in the critical load for zinc for forest systems related to eco-toxicological effects.

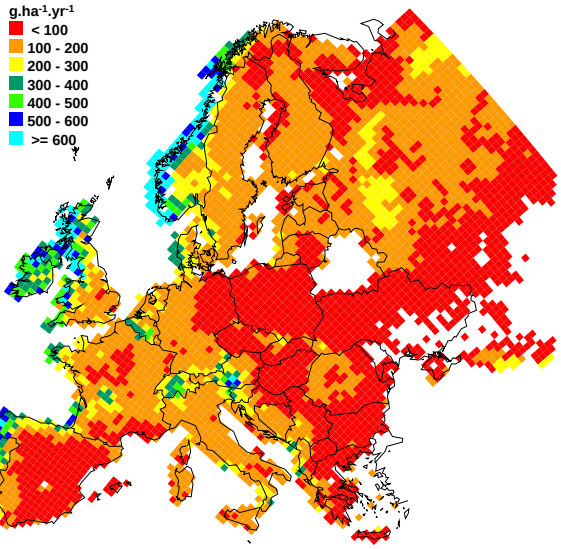



***Figure 3.7.2*** *Critical load for zinc for forests related to ecosystem functioning*

Critical loads related to ecosystem functioning, range from 70 - 700 $g.ha^{-1}.yr^{-1}$. Critical concentrations were computed as a function of DOC and pH and vary between 20 and 90 $\mu g\ l^{-1}$. For the lowest critical loads (< 80 $g.ha^{-1}.yr^{-1}$ coinciding with areas with low leaching rates), about half of the critical load consist of leaching an the other half of uptake, so that apart from the spatial pattern in the leaching flux, also the patterns in forest growth and crop yield are reflected in the critical load pattern. Since these patterns are to some extend correlated, they cannot be easily distinguished on the map.

## 3.7.4 Exceedance of critical loads of zinc

Accumulated Average Exceedances for Zn have been calculated for human health, ecosystem and the minimum of both end points.

Zinc can be toxic for ecosystems. Figure 3.7.3 shows that the AAE for 2000 occur mainley in Poland and Ukraine. After implementation of FIAM the computed risk to ecosystems is negligible. Table 3.7.2 indicates that the risk to ecosystems in Europe covers less than 1% of the European ecosystem area in 2020.
However, it should be noted that deposition compuations of Zn are likely to be underestimated.

***Table 3.7.2*** *Percentage of the area at risk of health or environmental effects caused by deposition of zinc in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively.*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| AL | 8,730 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 56,364 | 0.03 | 0.09 | 0.09 | 0.03 |
| BA | 21,474 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 16,845 | 0.13 | 0.00 | 0.00 | 0.00 |
| BG | 85,024 | 0.95 | 0.04 | 0.04 | 0.00 |
| CH | 10,863 | 0.12 | 0.16 | 0.16 | 0.16 |
| CZ | 63,383 | 0.63 | 0.02 | 0.02 | 0.00 |
| DE | 258,260 | 2.48 | 3.88 | 3.88 | 2.15 |
| DK | 26,497 | 0.00 | 0.00 | 0.00 | 0.00 |
| EE | 28,567 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 367,032 | 1.36 | 0.13 | 0.13 | 0.00 |
| FI | 194,992 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 404,505 | 0.39 | 0.56 | 0.56 | 0.03 |
| GB | 63,064 | 0.02 | 0.00 | 0.00 | 0.00 |
| GR | 59,477 | 1.21 | 1.22 | 1.22 | 0.09 |
| HR | 12,525 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 75,044 | 0.20 | 0.03 | 0.03 | 0.00 |
| IE | 8,489 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 220,767 | 0.36 | 0.40 | 0.40 | 0.00 |
| LT | 49,547 | 0.00 | 0.04 | 0.04 | 0.00 |
| LU | 2,055 | 0.00 | 8.85 | 8.85 | 0.00 |
| LV | 37,623 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 16,586 | 0.29 | 0.32 | 0.32 | 0.00 |
| NO | 63,680 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 214,819 | 15.01 | 4.34 | 4.34 | 0.32 |
| PT | 51,767 | 0.00 | 0.02 | 0.02 | 0.00 |
| RO | 191,953 | 0.01 | 0.00 | 0.00 | 0.00 |
| RU | 1,032,918 | 0.52 | 0.76 | 0.76 | 0.00 |
| SE | 200,470 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 17,109 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 35,773 | 1.26 | 0.37 | 0.37 | 0.11 |
| UA | 77,792 | 14.03 | 11.49 | 11.49 | 0.02 |
| YU | 29,861 | 0.17 | 0.09 | 0.09 | 0.00 |
| EU25 | 2,460,867 | 1.94 | 0.98 | 0.98 | 0.26 |
| CLRTAP Europe | 4,003,856 | 1.66 | 1.06 | 1.06 | 0.16 |

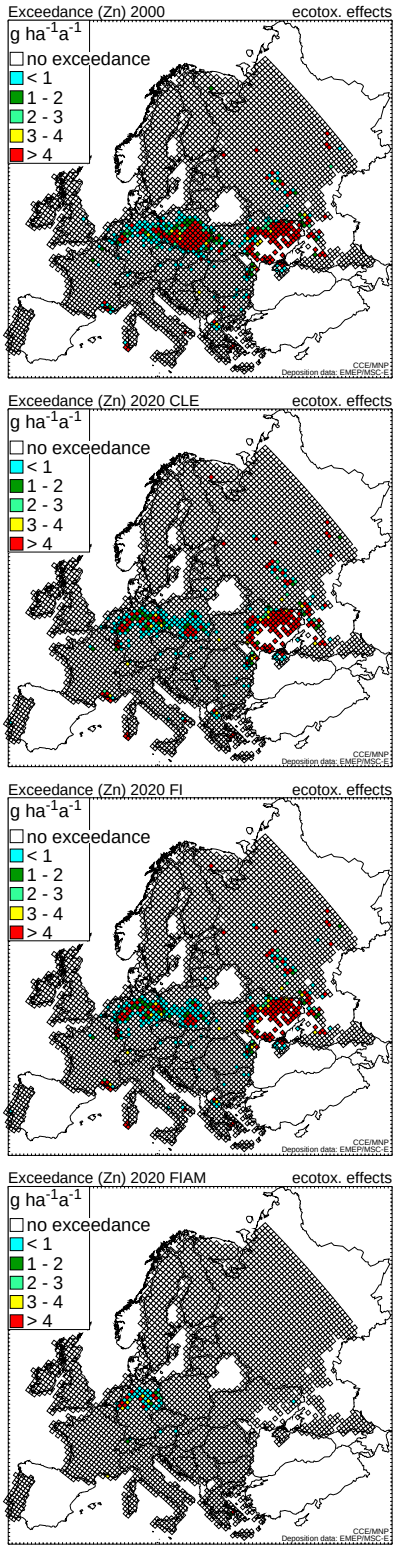



***Figure 3.7.3*** *Accumulated Average Exceedances of the critical loads of zinc with respect to ecosystem health in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4).*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

# 3.8. Arsenic

## 3.8.1. Emissions

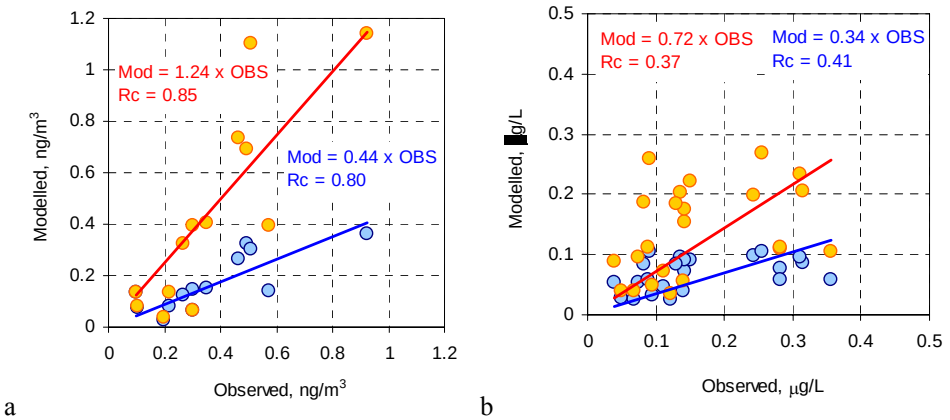
***Table 3.8.1*** *National arsenic emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 175 | 173 | 178 | 155 | 121 |
| Armenia | | 96 | 116 | 106 | 103 | 103 |
| Austria | | 2794 | 2665 | 2821 | 2821 | 2505 |
| Azerbaijan | | 2285 | 2753 | 2687 | 2684 | 208 |
| Belarus | | 3360 | 3623 | 3118 | 2616 | 1523 |
| Belgium | | 3840 | 1631 | 1490 | 1490 | 1231 |
| Bosnia-Herzegovina | | 2239 | 2121 | 1929 | 577 | 312 |
| Bulgaria | | 3474 | 1221 | 1011 | 1011 | 485 |
| Croatia | | 1068 | 629 | 435 | 379 | 164 |
| Cyprus | | 629 | 613 | 718 | 718 | 40 |
| Czech Republic | | 11634 | 2869 | 2348 | 2348 | 2178 |
| Denmark | | 974 | 696 | 470 | 470 | 455 |
| Estonia | | 9668 | 2801 | 1920 | 1920 | 1563 |
| Finland | | 4500 | 4040 | 4495 | 4495 | 3300 |
| France | | 25264 | 20439 | 20444 | 20444 | 16701 |
| Georgia | | 173 | 217 | 223 | 211 | 83 |
| Germany | | 34520 | 31602 | 33587 | 33587 | 30525 |
| Greece | | 3975 | 2462 | 2216 | 2216 | 1273 |
| Hungary | | 5717 | 3070 | 2698 | 2698 | 2124 |
| Iceland | | 94 | 86 | 73 | 73 | 58 |
| Ireland | | 1757 | 897 | 722 | 722 | 558 |
| Italy | | 42619 | 27197 | 25773 | 25773 | 20733 |
| Kazakhstan | | 43818 | 51027 | 54606 | 30134 | 13393 |
| Kyrgyzstan | | 478 | 577 | 526 | 286 | 246 |
| Latvia | | 624 | 618 | 671 | 671 | 400 |
| Lithuania | | 792 | 972 | 811 | 811 | 728 |
| Luxembourg | | 79 | 57 | 53 | 53 | 52 |
| Macedonia (Form. Yug. Rep. of) | | 760 | 667 | 572 | 345 | 153 |
| Moldova (Rep. of) | | 477 | 395 | 239 | 239 | 215 |
| Netherlands | | 1258 | 1002 | 960 | 960 | 890 |
| Norway | | 2457 | 2268 | 2291 | 2291 | 1841 |
| Poland | | 50400 | 22617 | 16508 | 16508 | 14752 |
| Portugal | | 4515 | 2469 | 2515 | 2515 | 1638 |
| Romania | | 4602 | 2292 | 2058 | 2058 | 624 |
| Russia | | 125475 | 146926 | 154238 | 82677 | 33715 |
| Slovak Republic | | 11219 | 4280 | 3156 | 3156 | 3032 |
| Slovenia | | 789 | 311 | 304 | 304 | 256 |
| Spain | | 56072 | 27854 | 18488 | 18488 | 16805 |
| Sweden | | 594 | 583 | 879 | 879 | 739 |
| Switzerland | | 798 | 734 | 750 | 750 | 668 |
| Turkey | | 15463 | 15222 | 15318 | 9864 | 5825 |
| Ukraine | | 30334 | 31821 | 30828 | 17682 | 12556 |
| United Kingdom | | 38022 | 19480 | 17929 | 17929 | 15576 |
| Yugoslavia (Fed. Rep. of) | | 5168 | 4974 | 4646 | 2026 | 840 |
| ***Total (tonnes/yr)*** | | ***555*** | ***449*** | ***438*** | ***318*** | ***211*** |

## 3.8.2 Depositions

Depositions for As are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions. The calculated As depositions are most probably an underestimation.

Air concentrations of arsenic based on official/TNO emissions for 2000 are underestimated 2 - 2.5 times (Figure 3.8.1a.). Similar degree of discrepancy took place for concentrations in precipitation (Figure 3.8.1b.). This underestimation means that modelled depositions tend to be lower than "real" ones. Therefore, "real" area of regions where depositions are close or exceed a critical load value may be higher than that derived from modelled depositions. Coefficient of correlation for air concentrations is significant (0.8), which imply that the model captures the spatial pattern of air concentrations. Therefore, the underestimation of the observed values may be connected with insufficient atmospheric emissions (anthropogenic or natural and historic). The use of ESPREME emissions gives better agreement between modelled and measured air concentrations as well as concentrations in precipitation. Correlation coefficient for concentrations in precipitation is quite low for both emission data sets. This may indicate at the necessity of revision of wet scavenging parameters for arsenic. Another reason may be connected with quality of measurement data.




a b

***Figure 3.8.1*** *Comparison of modelled and measured arsenic concentrations in air (a) and concentrations in precipitation (b), using official/ TNO (blue) and ESPREME (red) emissions. Natural and historical emissions are included. Rc = correlation coefficient*

## 3.8.3 Critical loads

Figure 3.8.2 shows the regional pattern in the critical load for arsenic for agricultural systems related to human health effects.

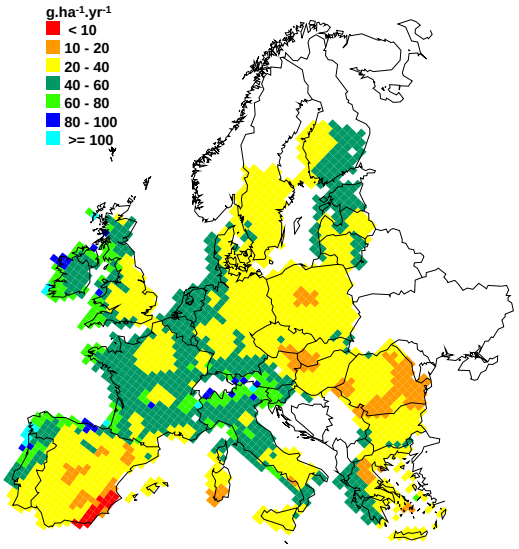



***Figure 3.8.2*** *Critical load of As for agriculture related to human health effects.*

The critical concentration for human health effects was set at 10 $mg.m^{-3}$ which is much lower than the limit of 70 $mg.m^{-3}$ related to eco-toxicological effects. As a consequence, critical loads for arsenic related to human health effects are much lower than those for eco-toxicological effects: critical loads for public health vary between 5 - 70 $g.ha^{-1}.yr^{-1}$, those for eco-toxicology vary between 40 - 400 $g.ha^{-1}.yr^{-1}$. Because the arsenic contents in crops and trees are very low (0.1 and 0.02 $mg.kg^{-1}$ respectively), uptake does hardly contribute to the critical load.

## 3.8.4 Exceedance of critical loads of arsenic

Accumulated Average Exceedances for As have been calculated for human health, ecosystem and the minimum of both endpoints.

Both Table 3.8.2 and Figure 3.8.3 indicate that there is hardly any (except for 0.08% of Polish ecosystems in 2000) risk of adverse effects caused by the long-range atmospheric deposition of arsenic. However, it should be noted that deposition computations of As are likely to be underestimated.

***Table 3.8.2**: Percentage of the area at risk of health or environmental effects caused by deposition of arsenic in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively.*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| Al | 17,459 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 112,715 | 0.00 | 0.00 | 0.00 | 0.00 |
| BA | 42,948 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 33,690 | 0.00 | 0.00 | 0.00 | 0.00 |
| BG | 170,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| CH | 21,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| CZ | 126,757 | 0.00 | 0.00 | 0.00 | 0.00 |
| DE | 516,512 | 0.00 | 0.00 | 0.00 | 0.00 |
| DK | 52,987 | 0.00 | 0.00 | 0.00 | 0.00 |
| EE | 57,134 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 734,064 | 0.00 | 0.00 | 0.00 | 0.00 |
| FI | 389,983 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 808,993 | 0.00 | 0.00 | 0.00 | 0.00 |
| GB | 126,106 | 0.00 | 0.00 | 0.00 | 0.00 |
| GR | 118,953 | 0.00 | 0.00 | 0.00 | 0.00 |
| HR | 25,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 150,082 | 0.00 | 0.00 | 0.00 | 0.00 |
| IE | 16,978 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 441,533 | 0.00 | 0.00 | 0.00 | 0.00 |
| LT | 99,071 | 0.00 | 0.00 | 0.00 | 0.00 |
| LU | 4,111 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 75,240 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 33,171 | 0.00 | 0.00 | 0.00 | 0.00 |
| NO | 127,361 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 429,639 | 0.08 | 0.00 | 0.00 | 0.00 |
| PT | 103,526 | 0.00 | 0.00 | 0.00 | 0.00 |
| RO | 383,891 | 0.00 | 0.00 | 0.00 | 0.00 |
| RU | 2,065,805 | 0.00 | 0.00 | 0.00 | 0.00 |
| SE | 400,937 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 34,201 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 71,547 | 0.00 | 0.00 | 0.00 | 0.00 |
| UA | 155,570 | 0.00 | 0.00 | 0.00 | 0.00 |
| YU | 59,711 | 0.00 | 0.00 | 0.00 | 0.00 |
| EU25 | 4,921,598 | 0.01 | 0.00 | 0.00 | 0.00 |
| CLRTAP Europe | 8,007,488 | 0.00 | 0.00 | 0.00 | 0.00 |

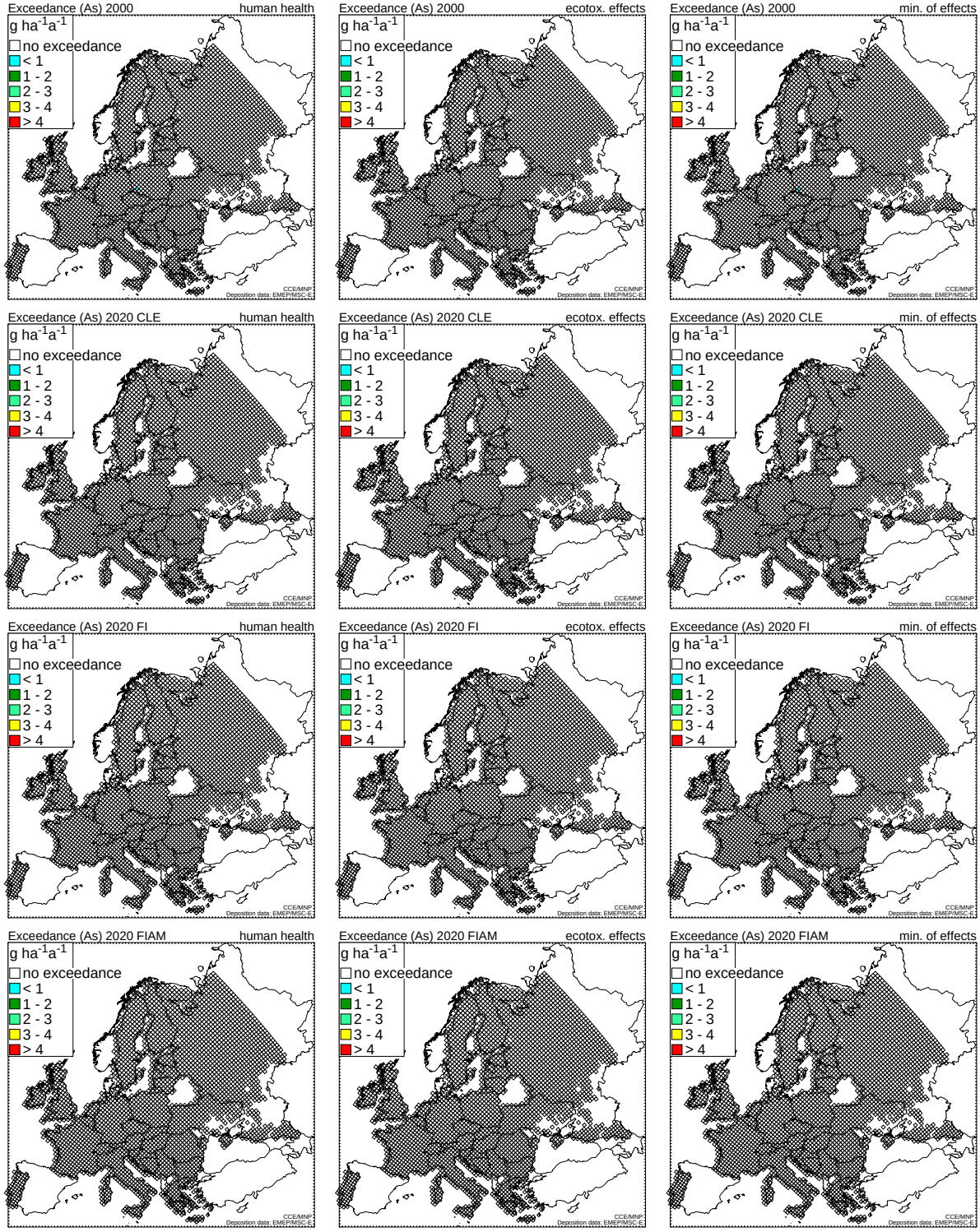



***Figure 3.8.3*** *Accumulated Average Exceedances of the critical loads of arsenic with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4).*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

# 3.9 Selenium

## 3.9.1 Emissions

***Table 3.9.1*** *National selenium emissions (kg/yr) for 2000 and projected emissions for 2010 and 2020 following Current legislation (CLE), Full implementation (FI) and FI+additional measures (FIAM ) scenarios*

| **Country** | **scenario** | | **CLE** | | **FI** | **FIAM** |
|---|---|---|---|---|---|---|
| | **year** | **2000** | **2010** | **2020** | **2020** | **2020** |
| Albania | | 127 | 125 | 125 | 117 | 94 |
| Armenia | | 153 | 166 | 154 | 154 | 109 |
| Austria | | 1826 | 1398 | 1481 | 1481 | 1239 |
| Azerbaijan | | 2043 | 2441 | 2377 | 2377 | 158 |
| Belarus | | 5462 | 5545 | 5397 | 5344 | 1891 |
| Belgium | | 4320 | 2282 | 2114 | 2114 | 1530 |
| Bosnia-Herzegovina | | 883 | 862 | 804 | 314 | 160 |
| Bulgaria | | 12384 | 11811 | 11644 | 11644 | 4667 |
| Croatia | | 633 | 437 | 400 | 400 | 167 |
| Cyprus | | 521 | 526 | 615 | 615 | 18 |
| Czech Republic | | 15785 | 4600 | 4256 | 4256 | 4112 |
| Denmark | | 2050 | 1352 | 1056 | 1056 | 1051 |
| Estonia | | 996 | 366 | 323 | 323 | 299 |
| Finland | | 5491 | 3218 | 3711 | 3711 | 3568 |
| France | | 14262 | 8508 | 8530 | 8530 | 6643 |
| Georgia | | 129 | 158 | 154 | 150 | 38 |
| Germany | | 30299 | 21138 | 21643 | 21643 | 20758 |
| Greece | | 1965 | 1504 | 1311 | 1311 | 481 |
| Hungary | | 1621 | 575 | 475 | 475 | 386 |
| Iceland | | 51 | 51 | 45 | 44 | 36 |
| Ireland | | 1414 | 678 | 497 | 497 | 349 |
| Italy | | 91478 | 31625 | 34293 | 34293 | 32878 |
| Kazakhstan | | 11253 | 12852 | 11714 | 6455 | 4358 |
| Kyrgyzstan | | 294 | 353 | 323 | 224 | 171 |
| Latvia | | 427 | 380 | 393 | 393 | 188 |
| Lithuania | | 1653 | 880 | 905 | 905 | 804 |
| Luxembourg | | 24 | 26 | 27 | 27 | 25 |
| Macedonia (Form. Yug. Rep. of) | | 237 | 186 | 139 | 96 | 35 |
| Moldova (Rep. of) | | 1707 | 1661 | 1654 | 1654 | 663 |
| Netherlands | | 2334 | 1405 | 1178 | 1178 | 1049 |
| Norway | | 496 | 513 | 516 | 516 | 442 |
| Poland | | 41845 | 16744 | 19498 | 19498 | 19090 |
| Portugal | | 24630 | 11468 | 15152 | 15152 | 14382 |
| Romania | | 9715 | 7741 | 7896 | 7896 | 2899 |
| Russia | | 74454 | 81750 | 80012 | 60986 | 24715 |
| Slovak Republic | | 7075 | 2414 | 2655 | 2655 | 2613 |
| Slovenia | | 404 | 195 | 191 | 191 | 149 |
| Spain | | 60781 | 25740 | 23936 | 23936 | 23593 |
| Sweden | | 569 | 463 | 911 | 911 | 858 |
| Switzerland | | 311 | 307 | 301 | 301 | 224 |
| Turkey | | 20775 | 23806 | 28762 | 27882 | 12442 |
| Ukraine | | 17224 | 17614 | 16821 | 11688 | 7531 |
| United Kingdom | | 28849 | 8725 | 8845 | 8845 | 8152 |
| Yugoslavia (Fed. Rep. of) | | 2372 | 2302 | 2240 | 2220 | 924 |
| ***Total (tonnes/yr)*** | | ***501*** | ***317*** | ***325*** | ***294*** | ***206*** |

## 3.9.2. Depositions

Depositions for Se are calculated for the 2000 official/TNO emissions and the three emission scenarios for 2020, all including natural and historic emissions.

Measurements of selenium at EMEP stations were not available for 2000. At station DK3 selenium concentrations in air were measured from 1979 to 1996, and at station IS91 – from 1995 to 1997 (Figure 3.9.1). Averaged concentrations at these stations are 0.61 and 0.18 ng/m$^3$, respectively. Modelled concentrations in 2000 were 0.13 ng/m$^3$ at DK3 and 0.02 ng/m$^3$ at IS91. Concentrations in precipitation have not been measured at all at EMEP network. As the measurements are almost absent, it is not possible to evaluate the model performance for this metal. Modelled concentrations in precipitation at monitoring stations ranged from 6 to 80 ng/L. In order to increase measurement database to validate the modelling results for selenium, the data from other sources, e.g., results of other monitoring programs, should be used.

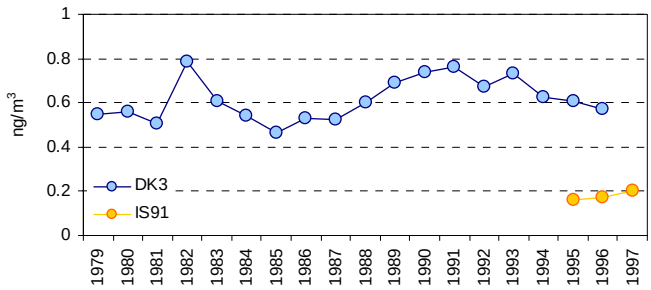



***Figure 3.9.1.*** *Measured concentrations of selenium at EMEP stations DK3 and IS91*

## 3.9.3. Critical loads

Figure 3.9.2 shows the regional pattern in the critical load for chromium for forest systems related to eco-toxicological effects.

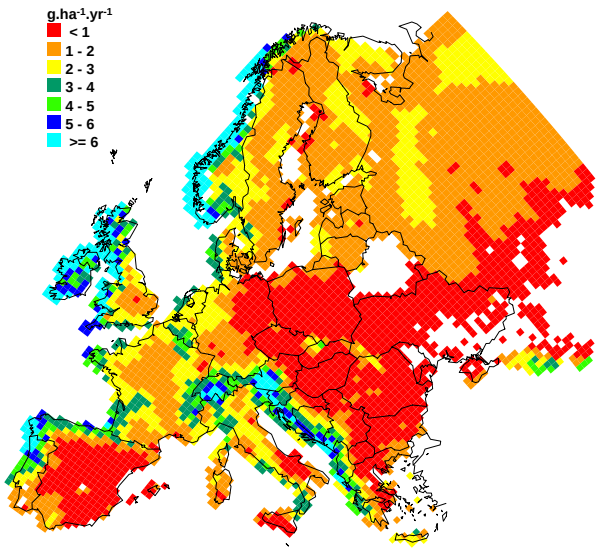



***Figure 3.9.2*** *Critical load of selenium for forests related to ecosystem functioning.*

For selenium a critical concentration related to ecosystem protection of 1 $mg.m^{-3}$ was used. For drinking water, the critical concentration was ten times higher: 10 $mg.m^{-3}$. This is directly reflected in the critical loads: critical loads for drinking water range from 5 - 70 $g.ha^{-1}.yr^{-1}$ whereas critical loads for ecosystem protection range from 0.5 - 6 $g.ha^{-1}.yr^{-1}$. As with the other metals, patterns in critical loads follow the patterns in precipitation excess. Uptake of selenium is generally low; only with the lowest eco-toxicological critical loads, uptake accounts for about 30 % of the critical load.

## 3.9.4. Exceedance of critical loads of selenium

Accumulated Average Exceedances for Se have been calculated for human health, ecosystem and the minimum of both endpoints.

Table 3.9.2 indicates that relatively large areas at risk in 2000 occur in the Ukraine (14%), Poland (13), The Czech Republic (10%), Slovakia (9%), Bulgaria (7%), Spain (7%), Germany (4%) and Hungary (2%).

The computed risk covers a reduced area in 2020-FIAM compared to 2000, but remains in Poland in particular (see Figure 3.9.3 bottom right map). Overall it can be concluded that the relative risk of impacts - i.e. the risk in 2020 compared to 2000 – can be reduced in Europe from about 2% in 2000 to below 1% in 2020.
However, it should be noted that emission data are uncertain and that monitoring data to compare with modelled data are scarce.

**Table 3.9.2** *Percentage of the area at risk of health or environmental effects caused by deposition of selenium in 2000, and in 2020 according to Current Legislation (CLE), Full Implementation (FI) and Full Implementation plus additional measures (FIAM) respectively.*

| Country | Area ($km^2$) | 2000 (%) | 2020 CLE (%) | 2020 FI (%) | 2020 FIAM (%) |
|---|---|---|---|---|---|
| Al | 17,459 | 0.00 | 0.00 | 0.00 | 0.00 |
| AT | 112,715 | 0.02 | 0.00 | 0.00 | 0.00 |
| BA | 42,948 | 0.00 | 0.00 | 0.00 | 0.00 |
| BE | 33,690 | 0.00 | 0.00 | 0.00 | 0.00 |
| BG | 170,043 | 7.42 | 7.02 | 6.93 | 0.38 |
| CH | 21,726 | 0.00 | 0.00 | 0.00 | 0.00 |
| CZ | 126,757 | 10.29 | 2.46 | 2.31 | 0.04 |
| DE | 516,512 | 3.78 | 1.04 | 1.03 | 0.06 |
| DK | 52,987 | 0.01 | 0.00 | 0.00 | 0.00 |
| EE | 57,134 | 0.00 | 0.00 | 0.00 | 0.00 |
| ES | 734,064 | 6.67 | 0.38 | 0.38 | 0.35 |
| FI | 389,983 | 0.00 | 0.00 | 0.00 | 0.00 |
| FR | 808,993 | 0.05 | 0.01 | 0.01 | 0.00 |
| GB | 126,106 | 0.72 | 0.00 | 0.00 | 0.00 |
| GR | 118,953 | 0.00 | 0.26 | 0.08 | 0.00 |
| HR | 25,043 | 0.00 | 0.00 | 0.00 | 0.00 |
| HU | 150,082 | 1.88 | 0.02 | 0.02 | 0.00 |
| IE | 16,978 | 0.00 | 0.00 | 0.00 | 0.00 |
| IT | 441,533 | 0.00 | 0.13 | 0.13 | 0.05 |
| LT | 99,071 | 0.05 | 0.00 | 0.00 | 0.00 |
| LU | 4,111 | 0.00 | 0.00 | 0.00 | 0.00 |
| LV | 75,240 | 0.00 | 0.00 | 0.00 | 0.00 |
| NL | 33,171 | 0.10 | 0.00 | 0.00 | 0.00 |
| NO | 127,361 | 0.00 | 0.00 | 0.00 | 0.00 |
| PL | 429,639 | 12.97 | 9.01 | 8.97 | 5.12 |
| PT | 103,526 | 0.71 | 0.17 | 0.17 | 0.12 |
| RO | 383,891 | 0.08 | 0.10 | 0.10 | 0.00 |
| RU | 2,065,805 | 0.48 | 0.56 | 0.25 | 0.00 |
| SE | 400,937 | 0.00 | 0.00 | 0.00 | 0.00 |
| SI | 34,201 | 0.00 | 0.00 | 0.00 | 0.00 |
| SK | 71,547 | 9.38 | 1.98 | 1.97 | 0.13 |
| UA | 155,570 | 14.08 | 6.82 | 3.95 | 0.48 |
| YU | 59,711 | 0.00 | 0.00 | 0.00 | 0.00 |
| EU25 | 4,823,597 | 3.08 | 1.09 | 1.07 | 0.52 |
| CLRTAP Europe | 8,007,489 | 2.41 | 1.09 | 0.94 | 0.33 |

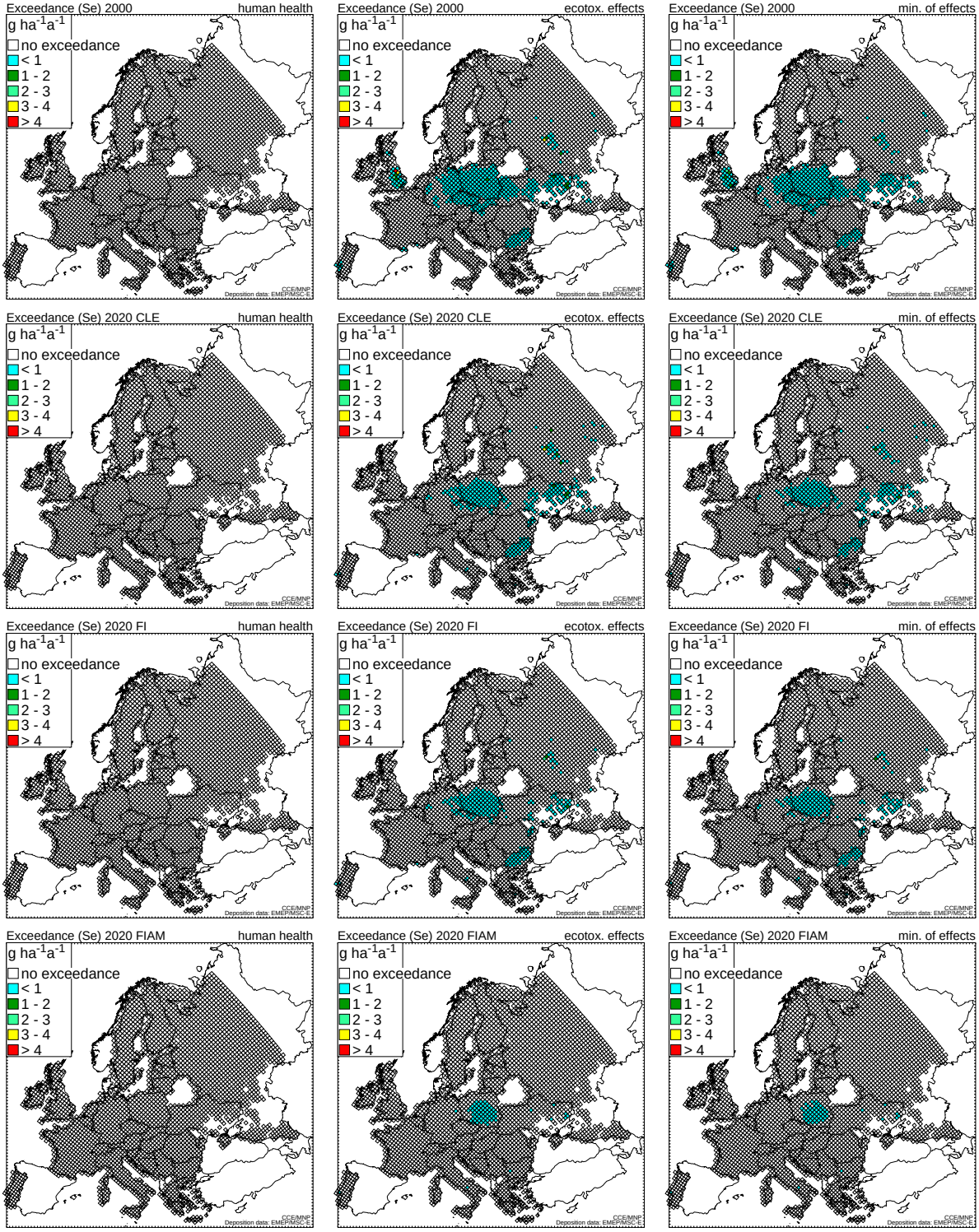



***Figure 3.9.3*** *Accumulated Average Exceedances of the critical loads of selenium with respect to human health (column 1), ecosystem (column 2) and the minimum of both endpoints (column 3) in 2000 (row 1) and in 2020 following CLE (row 2), Full Implementation ( row 3) and FI + Additional Measures (row 4).*
*Please note that exceedances in Belarus and parts of the Ukraine are not computed because forestry data is lacking to enable the computation of critical loads. The background database is not applied to compute exceedances for ecosystems in Denmark, Norway and Spain, similarly to agreed practice with respect to the priority metals.*

## References

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# 4. Conclusions

The critical loads of the 3 priority metals Hg (77%) , Pb (42%) and Cd are exceeded in 2000 leaving 77 %, 42 % and 34 % of the European ecosystem area at risk respectively. Full implementation of the HM Protocol in 2020 leaves more than 70% of the area at risk of mercury deposition (gaseous Hg is not addressed in the HM Protocol), 32% at risk of lead deposition and below 1% at risk of cadmium.

Possible additional measures on top of the full implementation of the HM Protocol further reduces the area with Pb exceedance to 19% and with Hg exceedance to 73%. Although the area of exceedance for Hg is only reduced with about 6 %-points in comparison to 2020-FI the Accumulated Average Exceedance over all European ecosystems in the EMEP domain is reduced from 0.120 to 0.064 g $ha^{-1}$ $a^{-1}$

The conclusions with respect to priority metals are in line with the Sufficiency and Effectiveness report of the Task Force on Heavy Metals and the findings of the TF on Health (Report on "Health Risks of HM from LRTAP", 2006)

The deposition of the other 6 HM in 2000 causes areas at risk in 2000 of less than 1%, except for the deposition of Zn and Se putting about 1.7% and 2.4% of the area at risk in 2000. Full implementation of the HM Protocol decreases this exceedance to about 1%. Additional measures on top of the HM Protocol reduces the area at risk further to well below 1%. Regarding 'other metals' the Sufficiency and Effectiveness report indicates that LRTAP dispersion is unlikely to reach high enough concentrations as a result of LRTAP to cause adverse effects on wildlife and human health.

Conclusions on exceedances should be handled with care:

-Computed current and projected future depositions for most of the heavy metals are likely to be underestimated. Especially for 2000, the use of officially reported emissions as well as TNO estimations leads to modelled depositions that are an underestimation of actual depositions in that year. Only for Hg a good match exists between calculated and observed data. For Pb and Ni the underestimation is around 30-40% and the other HM are underestimated by 2-3 times except for Zn of which the deposition could be underestimated by up to an order of magnitude. The tentative use of higher emission data by EMEP-MSCE (see ESPREME project) demonstrated that an improved agreement between modelled and measured quantities in 2000 can be obtained.

-Critical loads for the other 6 HM have been derived for the first time and have not been thoroughly reviewed yet, as has been the case for the critical loads of the 3 priority HM.

-Other loads not coming from air pollution, e.g. Cd, Cu, Pb and Zn from fertilisers and manure have not been taken into account when calculating exceedances.

The final conclusion is that the policy focus on the three priority metals Cd, Hg and Pb is justified.