

# **Methods to Analyze Interactions between Emissions of Air Pollutants in Europe**

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## ABSTRACT

This paper summarizes the results of a study on characteristics and requirements of emission inventories for Europe, to be used in integrated assessments that simultaneously analyze future global warming, acidification, eutrophication and ozone related problems. Such simultaneous analyses may avoid sub-optimal policy recommendations. We include emissions of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and fluorinated compounds (CFCs, halons, HCFCs, HFCs and SF<sub>6</sub>) nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>), ammonia (NH<sub>3</sub>), volatile organic compounds (VOC) and soot in our analysis. We discuss different types of interactions between emissions of air pollutants, and their underlying sources. Existing emission inventories for Europe do not account for all interactions and differ with respect to pollutants included, system boundaries, aggregation level, sources of emissions included, uncertainty assessment and underlying method for estimating emissions. Also environmental models differ with respect to reduction strategies considered. We discuss requirements of emission inventories and to what extent available inventories meet these requirements.

## 1 INTRODUCTION

Air pollution problems such as global warming, acidification, eutrophication, and ozone related problems are linked in many ways. Many human activities contribute to more than one environmental problem. Environmental policies, however, typically focus on just one environmental issue and tend to ignore side-effects on other pollutants. To develop more integrated solutions to air pollution problems, we need to consider possible interactions between these problems. Integrated analysis of air pollution problems requires a complex system that considers linkages between and within economic sectors, atmospheric processes, the environmental impact and possible policy responses. As yet, there is no integrated assessment modeling system available that simultaneously deals with global warming, acidification, eutrophication and ozone related problems. As a first step towards the development of such a system, it is worthwhile to study interactions that relate to *emissions* of various air pollutants.

This paper summarizes some important results of an analysis of interactions between emissions of air pollutants in Europe. A full report on this study can be found in [1]. The purpose of this study was to analyze some basic characteristics and requirements of emission inventories for Europe to be used in integrated assessments of future global warming, acidification, eutrophication and ozone related problems. Such analyses need to consider some important interactions between these air pollution problems, including interactions between economic sectors, atmospheric processes, environmental impact and policy options. We focus on European emissions of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and fluorinated compounds (CFCs, halons, HCFCs, HFCs and SF<sub>6</sub>) nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>), ammonia (NH<sub>3</sub>), volatile organic compounds (VOC) and soot.

## 2 LINKAGES BETWEEN SOURCES OF POLLUTANTS AND ENVIRONMENTAL PROBLEMS

We first answered the question of what the most important interactions are between emissions of air pollutants and the underlying sources. Our analysis shows that there are at least four types of interactions between economic sectors, atmospheric processes and environmental impacts that affect emissions of air pollutants.

(a) *Human activities giving rise to emissions of more than one pollutant*

Economic activities result usually in emissions of more than one gas. Moreover, emissions of pollutants can result from more than one economic activity. For instance, the combustion of energy, the industrial sector and road transport are important sources of major gases contributing to climate change, acidification as well as tropospheric ozone formation [2].

*(b) Biogenic and biogeochemical processes underlying emissions of more than one gas*

Several air pollutants are produced through biogenic and biogeochemical processes. These processes produce a number of gases and influence the concentrations of several pollutants. Only fluorinated compounds and soot are not a product of biogenic or biogeochemical processes. Most of these processes take place in soils and aquatic systems. Although these are natural processes, they are affected by human activities, in particular agriculture. Certain compounds, for instance  $\text{NO}_x$  and  $\text{N}_2\text{O}$  are emitted directly from the agricultural soils due to nitrification or denitrification, which is enhanced by fertilizer use. Anthropogenic deposition of N- compounds can result in indirect formation of  $\text{NO}_x$  and  $\text{N}_2\text{O}$  through enhanced denitrification in natural soils. This way, the emissions of  $\text{NO}_x$  and  $\text{NH}_3$  or other nitrogen compounds can be deposited on soils and oceans, and they can be re-emitted as  $\text{N}_2\text{O}$  into the atmosphere [3].

*(c) Impact of reduction strategies affecting more than one gas*

Reduction strategies for air pollution are designed to affect targeted emissions, but may in some cases as a side-effect affect emissions of other compounds. For instance, a possible reduction strategy for  $\text{SO}_2$  and  $\text{NO}_x$  emissions is a fuel switch from coal to natural gas. This reduces emission of  $\text{SO}_2$  and  $\text{NO}_x$ . Thus, this fuel switch reduces acidification. As a side-effect it may also lead to lower emissions of mercury and other hazardous air pollutants, including particulate matter [4]. In other cases, lowering emissions of one gas may increase emissions of other gases. For example, some technical measures to reduce  $\text{NH}_3$  emissions from manure may increase  $\text{N}_2\text{O}$  or nitrate leaching [5]. These examples show that it is important to consider the whole chain of interactions between the components before a new mitigation option is applied.

*(d) Effects of changes in the environment on emissions of air pollutants*

Changes in the environment can influence emissions of air pollutants, in particular when biogeochemical processes are affected. Those processes are typically influenced by environmental factors such as temperature, precipitation and acidity. Changes in one of these may therefore affect biogenic emission. This impact is often difficult to show, due to complexity of atmospheric or biogeochemical reactions. Sometimes the impact is not direct and requires studies of chains of events. But there are no doubts that such impacts exist. For instance, Smith et al. [6] show that changes in ultraviolet radiation due to stratospheric ozone depletion over Antarctica, have a direct impact on the phytoplankton production in water. They show that the increased UV-B radiation is causing a 6-12% decrease in phytoplankton productivity in Antarctic waters. As phytoplankton forms a large sink for atmospheric  $\text{CO}_2$ , this may influence atmospheric  $\text{CO}_2$  concentration. Another example is the potential effect of global warming on microbial  $\text{N}_2\text{O}$  production. Secondary effects of environmental change may also occur through changes in economic activities. As a result of global warming, power plants may reduce their production in wintertime, because demand for space heating will be smaller.

### **3 EMISSION DATABASES**

Next, we described characteristics of existing emission databases with respect to the compounds included, spatial and temporal specification, source categories included, and methods used to estimate emissions. We compared a selection of emission inventory databases and emission databases that are used as input to atmospheric models.

We included a selection of existing emission inventory databases in our comparison. One of them is the Emission Database for Atmospheric Research (EDGAR 2.0) [7], which is a global scientific database including emissions of greenhouse gases and a number of other compounds. We also included national emission inventories from databases of the European Monitoring and Emissions Program (EMEP/CORINAIR) [8], [9] and for the UN Framework Convention on Climate Change (UNFCCC) [10]. The Pollutant Emission Register (PER) [11] from The Netherlands is also included as an example of a detailed national emission inventory. Integrated Assessment models like the European air pollution model RAINS 7.2 [12], [13] and the climate model IMAGE 2.1 [14] use emission databases as input, as do atmospheric transport models like LOTOS [15], [16].

In Table 1 we compare a few emission inventories and model databases. The comparison is focusing in particular on (a) the number of compounds included, (b) the source categories of emissions, (c) the spatial and temporal characteristics, (d) the assessment of uncertainties and (e) the method used in emission calculations.

From our comparison of databases we can observe the following:

- (a) Different emission databases differ with respect to the *number of compounds included*. Most inventories focus on certain groups of pollutants (e.g. acidifying gases, greenhouse gases, and heavy metals). None of the inventories, however, includes all gases that we consider important contributors to global warming, acidification, tropospheric ozone formation and stratospheric ozone depletion. Soot, for instance, is not included in any of these inventories. EMEP and CORINAIR inventories also do not include fluorinated compounds, while the RAINS and LOTOS emission databases are only for a limited number of transboundary air pollutants .
- (b) Another important feature of emission inventories is the list of *source categories* of emissions explicitly accounted for. The inventories compared here differ to a large extent with respect to the number of sources included. CORINAIR, for instance, uses a system that specifies 375 detailed source categories. In RAINS Europe 7.2, on the other hand the subdivision of energy-related emission sources is based on different types of fuel that are used in less than ten economic sectors, while emissions from agriculture are specified for a limited number of animal and fertilizer categories.
- (c) A third characteristic of emission databases relates to *spatial* and *temporal* aspects. The databases differ considerably in this respect. Most of the analyzed inventories cover Europe but for instance the database of IMAGE 2.0 covers the whole world. Most databases are based on national emission inventories. Nevertheless, the spatial aggregation level varies among the inventories. The global or continental inventories are mostly disaggregated into regions and countries. This holds for CORINAIR, IMAGE 2.0 and RAINS Europe 7.2. To use the inventory as a tool in integrated models, the estimates must be available in a comparable form. In many atmospheric transport models like LOTOS, on the other hand, the input must be given as emissions from all point and non-point sources per grid cell. The *temporal* characteristics of the emission inventories are different as well. EDGAR and LOTOS include only historical estimates, while RAINS and IMAGE also include projections for future emissions. In most of the analyzed cases the emission estimates are given as an annual total emission (CORINAIR, IMAGE 2.0, RAINS Europe 7.2). Some databases use additional models that include the climatic and seasonal trends to produce more detailed emission trends in time. This is an important aspect because atmospheric transport models like LOTOS need to process the data on a daily or even hourly basis.

- (d) An *uncertainty assessment* of the emission estimates has in most of the cases not been performed, or only to a limited extent. Available uncertainty assessments typically include partial quantitative analyses or a qualitative description of the quality of data.
- (e) Finally, we compared the *method* of emission estimation. In most databases a so-called 'emission factor approach' was followed, in which activity levels are multiplied by emission factors, that reflect the emissions per unit of activity. Differences between the emission databases can be attributed to differences in the estimation of activity levels and/or emission factors [17].

## 4 REDUCTION STRATEGIES

A next question that arises is what types of reduction strategies are typically included in available modeling studies of air pollution problems in Europe. To this end, we compared the following models IMAGE, RAINS, LOTOS, MERGE, and MARKAL.

The models that we included in our comparison differ from each other in many aspects. Some of the models are mainly used for scenario analysis of future developments, based on “what if..” type of questions. For instance **IMAGE 2.0** is an Integrated Assessment model for global climate change and mainly used for scenario analysis [14]. Also the **RAINS Europe 7.2**, a European air pollution model, can be used for scenario analysis, and contains three sets of reduction strategies for SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub> [13]. Other than in IMAGE 2.0, the RAINS reduction strategies are explicitly included in the model and grouped for individual air pollutants. Moreover, every option can be used separately or the user can define sets of them. RAINS includes information on the reduction potential of reduction strategies and on the costs of these technologies.

RAINS is not only used for scenario analyses, but also for determining cost-optimal strategies to reduce acidification and eutrophication (optimization analyses). An Asian version of RAINS has been released recently, which so far only includes SO<sub>2</sub>. As in RAINS Europe, the model includes end of pipe technologies for the reduction of sulfur emissions and their costs [18]. Other than in RAINS Europe, however, the model also includes fuel switch as an option to reduce emissions. It considers a number of renewable fuels that can avoid the building of new coal fired power plants that are envisaged in the baseline projections of the model [19]. When used in optimization mode to identify the most cost efficient ways to reduce emission, RAINS-Asia treats the fuel switches as if they were end of pipe technologies.

The RAINS approach is different from models like MERGE and MARKAL, that include a full energy model to determine the cost optimal fuel mix. **MERGE** (Model for Evaluating Regional and Global Effects versions 2-4) is a dynamic general equilibrium model that generates Pareto optimal paths of investment and (energy) production for five (Version 2) and nine (Version 3 and 4) world regions over more than 10 decades, given the following inputs: potential gross domestic product, population, and (energy) production technologies [20]. The model calculated energy-related emissions of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O [21]. The **MARKAL Matter** (MARKet ALlocation) model is a representation of the economy of a region, represented by processes and physical and monetary flows between these processes. Four greenhouse gases are considered: CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and PFC (perfluorocarbons). Emissions are valued in financial terms on the basis of CO<sub>2</sub> equivalent penalty level that is set by the user. **MARKAL MATTER** (MATERials Technologies for GHG Emission Reduction) is the latest version of MARKAL and it includes detailed abatement strategies. The model covers 25 energy carriers and 125 materials, and the following reduction strategies [22]: (a) industrial processes improvements, (b) CO<sub>2</sub> removal from industrial plants, (c) end of pipe technologies and process substitution, (d) reduction of material consumption through product

substitution, (e) materials substitution, (f) renewable biomass feedstocks, (g) improved waste collection and separation system, (h) waste recycling, cascading and energy recovery.

A comparison of these models reveals that these models differ with respect to the type of analysis for which they were designed, and this affects the way reduction strategies are included. IMAGE, for instance, does not explicitly account for reduction options in detail, but rather defines packages of options as part of more general reduction strategies. RAINS includes a wide range of technical reduction options for transboundary air pollutants. However, RAINS largely focuses on technical end-of-pipe technologies. Models like MERGE and MARKAL include more complete energy modules and can be used to identify the cost optimal fuel mix. These models, however, usually do not include all technical options to reduce emissions other than greenhouse gases. Atmospheric transport models like LOTOS [15] are developed for modeling atmospheric processes and do not include reduction strategies at all.

We compared the models also with respect to reduction options for specific economic sectors. In our comparison we distinguished between two types of reduction strategies. Demand-side options typically reduce the demand for a certain product or activity. Supply-side options on the other hand, reduce emissions per unit of product or activity.

For the *energy sector* the reduction options, on the demand side, are reducing the demand for energy and therefore affect the end use of energy. These options include energy efficiency improvement, as well as changes in human behavior. Supply side options reduce the emissions per unit of produced energy and can be subdivided into three groups. The first subgroup covers end of pipe technologies. To this group belong technologies that are used just at the end of the production process. RAINS Europe 7.2 includes end of pipe technologies to reduce SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub> emissions, while IMAGE 2.0 considers end of pipe technologies to mitigate greenhouse gases. The second subgroup of supply side strategies is based on fuel switch. In the RAINS Europe 7.2 model, a limited fuel switch is possible: regular petrol can be replaced by low sulfur petrol. In models like MERGE and MARKAL the energy system is modeled in detail to analyze the optimal fuel mix. To the third subgroup belong options that improve the efficiencies of electricity and heat production and hence reduce the emissions from power plants and heat production units. These types of options are usually not explicitly modeled in existing models.

For the *industrial sector* the demand side options include strategies to affect the demand for the products of industry. Of the examined models only MERGE includes such options. Alternatively, options might reduce the emissions per unit production. These are grouped in three categories: (i) end of pipe technologies, (ii) change in production processes and (iii) efficiency improvement. The first group consists of options that are used in the last stage of production processes. Changes in production processes concern process input technologies in order to reduce emissions. Efficiency improvement options are those ones, that are used in the production process in order to minimize energy use or to optimize production processes. Of the analyzed models IMAGE 2.0 includes these options but it is restricted to carbon dioxide emissions reduction.

For *agricultural* emissions there are also many abatement options available. Demand side options may change the need for certain agriculture products. Supply side consists of three categories: (i) technological options, (ii) changes in production processes and (iii) efficiency improvement. The first group contains technologies that reduce the emissions at the end of the production processes. RAINS Europe 7.2 and IMAGE 2.0 include those options. The changes of production technologies are in the second group. Such options for reducing N<sub>2</sub>O emissions are included in IMAGE 2.0. Efficiency improvement options, the third group, comprise for instance efficiency improvement of fertilizers.

We conclude that the models usually do not simultaneously include demand and supply side options for a wide range of gases and economic sectors. The reduction strategies are typically designed for the model's particular purposes and usually deal with one environmental problem only. In most of the cases models do not include the interactions between different pollutants. Therefore the emissions of not-targeted gases can be influenced by abatement strategies. In none of the analyzed models the reduction strategies for ozone-depleting substances are included.

## **5 EMISSION INVENTORIES TO BE USED IN INTEGRATED ANALYSES**

So far, we discussed characteristics of existing emission databases and models. We showed that most databases and models focus on only one of the air pollution problems and do not account for interactions with other environmental problems. This raises the question of whether we can identify the basic requirements of emission inventories to be used in an integrated analysis of global warming, acidification, eutrophication and ozone-related problems in Europe.

We described the characteristics of an ideal emission inventory. These characteristics are derived from the research objectives of different types of studies (economic, atmospheric and policy analyses), and refer to the compounds included, spatial and temporal detail, source categories included, uncertainties and reduction strategies. Clearly, the requirements of emission inventories for use in economic models, atmospheric models and policy-oriented Integrated Assessment models differ largely. A combination of these requirements may reveal an “ideal” specification of emission inventory databases that would meet the needs of all three types of scientists and that would therefore be a good basis for fully integrated studies. Its spatial and temporal aggregation level would be highly detailed (gridded, 10' by 10'), to meet the needs of atmospheric transport and chemistry models. Atmospheric models would also require that the emissions are convertible into point and area sources. For policy analyses, it is desirable to include long term historical data sets as well as projections of future developments (at least several decades). Both for economic and policy analyses the database would explicitly account for a large number of source categories as well as options to reduce these emissions and their associated costs. Finally the uncertainties need to be accounted for.

An emission inventory database, which satisfies the requirements for economic, atmospheric and integrated assessment models may not be easy to realize. Such an “ideal” specification, is obviously a hypothetical case and not available at present for Europe. One of the largest obstacles in realizing detailed emission inventory databases is data availability. Some of the chemical compounds are not well documented and many have a high rate of uncertainty [23]. Another important reason is that it is difficult to obtain a dataset with a high temporal resolution. Most measuring stations do not measure in such detail, and once models are needed to convert data, the uncertainties are increasing. Apart from data collection, the relationship between activity of economic sectors, emissions and atmospheric processes are not known in all detail. Including interactions may further complicate these issues.

Table 2 shows that when comparing existing inventories and models to those ‘ideal characteristics’ we observe that none of the existing inventories meet these requirements. For instance, none of the inventories includes all compounds that we consider important, although the IMAGE database includes most of the gases, that are considered important. None of the databases is focusing on all four environmental problems that are studied here. With respect to the aggregation level, we show that the LOTOS database has the most detailed spatial and temporal specification, which is the closest to the ‘ideal’ spatial aggregation level, but only for a limited

number of gases and years without detailed specification of sources. With respect to the source categories included, IA models like RAINS and IMAGE are most detailed and include several economic sectors and abatement strategies. These models can also be used for analysis of future trends. However, IA models typically lack the temporal and spatial detail of atmospheric models like LOTOS. None of the existing models has been subject to systematic quantitative uncertainty analysis.

## **6 RECOMMENDATIONS FOR FUTURE RESEARCH**

Our analysis shows that for Europe a variety of emission inventory databases and model databases is available with varying characteristics. Yet, none of them allows for an integrated analysis of the air pollution problems that we focused on. We therefore recommend that future research activities explore the possibilities to link existing emission inventories in such a way that they can be considered standardized, consistent and complementary. Databases are not easily combined because of differences in sectoral, spatial and temporal specification. For instance, the definitions of source categories in EMEP/CORINAIR inventories differ from those in the National Communications for the UNFCCC, which again differ from those in EDGAR. Recent activities aimed at conversion of EMEP/CORINAIR inventories into the required specification for the UNFCCC and vice versa. It would be worthwhile to expand such efforts to other databases.

It would furthermore be helpful if emission inventory databases were made more flexible. Linking databases would be easier if databases could provide information at several levels of spatial, temporal and sectoral detail. EDGAR is an example of a database that can provide information on several spatial levels: gridded emission inventories for atmospheric models, and country- or regional inventories for economic or policy analyses. However, most emission databases cannot easily convert to other aggregation levels. Existing databases seem particularly poor in the ability to convert information into more detailed temporal scales (e.g. seasonal cycles), in combination with details on sources categories.

A first step towards a simultaneous analysis of different air pollution problems in Europe, could be to 'soft-link' existing models for the purpose of scenario analysis, addressing 'what if' type of questions. An example of this is the AIRCLIM project, where RAINS and IMAGE have been used to investigate linkages between regional air pollution in Europe (acidification mainly) and climate change. Including other air pollution problems may be a logical next step.

Another option could be to link models specifically designed for scenario analysis to models that have the possibility of optimization analysis or general equilibrium modeling. An example of this is the combination of the global climate model DIALOGUE (developed for scenario analysis with a focus on the impact of climate change) to MERGE (a climate model including a general equilibrium model of the energy-economic system) [24]. For analyses of air pollution problems in Europe, it would be interesting to seek for combinations of models like IMAGE or MERGE with RAINS or MARKAL. This linking may, however, lead to inconsistencies, which require special attention.

More long-term research goals could include the development of an integrated model that fully accounts for interactions between different air pollution problems in Europe. In this paper we described possible characteristics of emission databases that would be required for such a model. If we are able to develop an emission database that meets these needs, we can use this in the development of an integrated modeling framework. Such a framework would, ideally, provide the possibility to explore future trends in emissions as affected by human activities and their effects on

the environment. It should be developed such that it not only takes into account the interactions in the environmental system that affect emissions (as described in this paper), but also interactions within and between the economic system, the atmosphere and the natural environment. In order to study acidification, eutrophication, tropospheric ozone and climate change simultaneously, this modeling framework would require a detailed specification of all relevant air pollutants and their sources, an atmospheric dispersion model, emission reduction strategies and the associated costs. Including stratospheric ozone problems would not be our first priority, although there are several interesting interactions between ozone depletion and climate change. We recommend that for optimization analysis, objective functions and/or constraints be defined in terms of environmental impact. To this end, the integrated model should include sufficient environmental indicators such as CO<sub>2</sub>-equivalent emission targets for greenhouse gases, critical levels for tropospheric ozone concentrations, and critical loads for deposition of acidifying and eutrophying compounds.

**Key words:** emission inventories, model databases, greenhouse gases, reduction strategies, air pollution

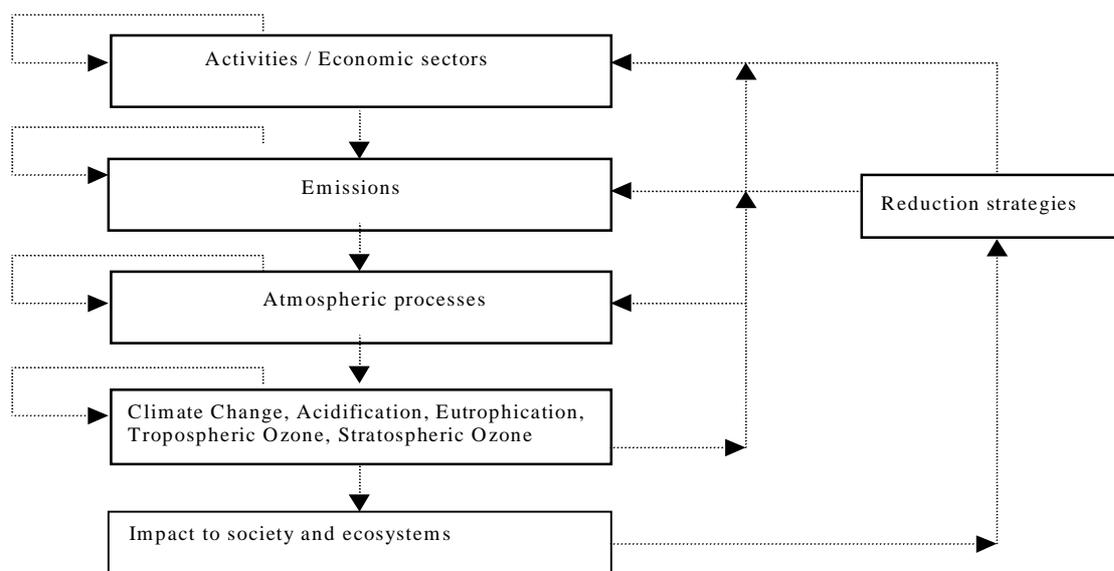
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**Figure 1**

Linkages between air pollution problems

**Table 1**

Comparison of a selection of emission inventories and model databases

|                                      | <i>Comparison</i>   |
|--------------------------------------|---|
| Number of compounds included         | Ranges from 1 (METDAT) to about 170 (PER)                     |
| Number of source categories included | Ranges from 12 (LOTOS) to 375 (CORINAIR)                      |
| Spatial system boundaries            | Range from Europe(CORINAIR) to World (IMAGE)                  |
| Spatial aggregation level            | Ranges from the country-level (CORINAIR) to fin grid (LOTOS)  |
| Temporal system boundaries           | Range from one year (LOTOS) to two centuries (IMAGE)          |
| Temporal aggregation level           | Ranges from annual totals (RAINS) to diurnal cycles (LOTOS)   |
| Uncertainty Assessment               | Limited in all databases                                      |
| Reduction strategies included        | Range from none (LOTOS) to selected sets of options in models |
| Methods of emission estimation       | In most cases an 'emission factor approach'                   |

**Table 2**

Comparisons of existing model databases to an 'ideal' specification of emission databases that allow for integrated analyses of different air pollution problems in Europe. + and – mean that model meet or do not meet ideal requirements, respectively.

|                            | <i>Ideal specification</i>          | <i>RAINS<br/>EUROPE 7.2</i> | <i>IMAGE 2.0</i> | <i>LOTOS</i> |
|----------------------------|-------------------------------------|-----------------------------|------------------|--------------|
| Components                 | All relevant compounds              | +/-                         | +/-              | +/-          |
| Spatial system boundaries  | Europe                              | +                           | +                | +            |
| Spatial aggregation level  | Detailed grid (10' x 10')           | -                           | -                | +            |
| Temporal system boundaries | Historic and long term trends       | +                           | +                | -            |
| Temporal aggregation level | Hourly averages                     | -                           | -                | +            |
| Emission sources included  | Detailed source categories          | +                           | +                | -            |
| Uncertainties              | Quantitative uncertainty assessment | +/-                         | -                | -            |
| Reduction strategies       | Demand- and supply-side options     | +                           | +                | -            |