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Towards an Integrated Assessment Model for Tropospheric Ozone- Emission Inventories, Scenarios and Emission-control Options

Olsthoorn, X.

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Working Paper

**Towards an integrated assessment
model for tropospheric ozone
Emission inventories, scenarios and
emission-control options**

Xander Olsthoorn

WP-94-27
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International Institute for Applied Systems Analysis A-2361 Laxenburg Austria

Telephone: +43 2236 715210 Telex: 079137 iiasa a Telefax: +43 2236 71313

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Preface

Ozone is a secondary pollutant, formed from emissions of nitrogen oxides, volatile organic compounds (VOC) and carbon monoxide. Due to the complex and non-linear photo-chemical atmospheric processes ozone concentrations can only be reduced by a balanced cut of the emissions of all three pollutants. The development of an ozone formation model is an important new activity of IIASA's Systems Analysis for Air Pollution project. The envisaged integrated assessment of ozone reduction strategies will have to combine information on the emission sources, the technical potential and costs of emission reduction measures as well as an understanding of the chemical processes influencing ozone formation. This paper reviews the available information material on VOC emissions, on their future development as well as the availability of measures and associated costs of controlling these emissions. Moreover, the paper proposes a design for a VOC emission model and its linkage to existing Europe-wide emission inventories.

Foreword

This paper is the result of the work done at the International Institute for Applied System Analysis (IIASA) during a three-month period in the spring of 1992. This stay was made possible through a grant from the Dutch NMO from a scheme aimed at promoting scientific collaboration between Dutch scientists and IIASA. I gratefully acknowledge the Dutch IIASA-NMO for providing the funds and the Institute for Environmental Studies of the Vrije Universiteit in Amsterdam for allowing the leave. Also I thank my temporary colleagues of the TAP project and IIASA staff for making my stay at IIASA both fruitful and pleasant.

Xander Olsthoorn

Abstract

IIASA intends to extend its RAINS model for addressing the issue of transboundary ozone air pollution. This requires the development of a VOC-emissions module, VOCs being precursors in ozone formation. The module should contain a Europe-wide emission inventory, a submodule for developing emission scenarios and a database of measures for VOC-emission control, including data about control effectiveness and control costs. It is recommended to use the forthcoming CORINAIR90 inventory for constructing the module. A proposal for a sectoral structure is made which is compatible with the RAINS and CORINAIR format. A preliminary database for VOC-abatement measures is presented.

1 INTRODUCTION

Since 1979, the year in which the UN-ECE (United Nations Economic Commission for Europe) adopted the Convention on Long-range Transboundary Air Pollution, the issue of trans-boundary air pollution due to ozone has attracted more and more attention. The international discussions focused on developing strategies to tackle this problem. At the end of 1991, an ECE protocol for the reduction of VOC (Volatile Organic Compounds) emissions was agreed on, VOC emissions being a main cause for ozone problems together with NO_x (nitrogen oxides). This agreement followed the protocol on the Reduction of Sulphur Emissions in 1985, and the 1988 protocol on the control of emissions of nitrogen oxides. This sequence of protocols reflects the development in the knowledge of the different air-pollution issues such as the emissions, atmospheric chemistry and transport and possibilities of emission control of the different chemical species.

This international development constitutes a background for strategies at the national levels and in the EC. Several European countries have adopted VOC policies and measures are already taken. The EC has adopted as objective in its fifth Environmental Action Programme to reduce man-made NMVOC emissions by 2000 with 30% of the 1990 level.

Integrated assessment models describe the entire cause-effect relationship of air pollution from the sources of emissions over atmospheric to environmental impacts. At the moment such integrated models address the acidification problem (e.g. the Regional Acidification Information and Simulation (RAINS) model developed by IIASA's Transboundary Air Pollution Project; see Alcamo et al., 1990). The RAINS model is being used for scientific support of the international negotiations on emission reductions currently taking place under the framework of the UN-ECE Convention on Long Range Transboundary Air Pollution (Amann et al., 1991). In designing cost-effective strategies to resolve the problem of acidification the RAINS model has proved itself as a successful tool in the international coordination of the air-pollution policies of the various ECE countries. Obviously, this prompts the question whether a similar model can be constructed which can be useful in addressing the problem of trans-boundary ozone pollution.

Since the RAINS model contains already elements to analyze the role of nitrogen oxides emissions - in studying acidification -, it seems promising to extend the model to study of effects on tropospheric ozone formation. Although it is well known that the ozone issue is more complicated than the problem of acidification, in particular due to the chemistry involved, it was felt that currently such a model can be constructed. This expectation is based on:

- the experience and expertise obtained by developing and applying the RAINS model;
- the progress in modelling long-range photochemical air pollution;
- the current state-of-the-art of computing.

An additional incentive for this study is the prospect to explore efficient - integrated - strategies to reduce environmental damage due to both acidification and ozone formation. Such an extension requires the following steps:

- Analysis of VOC emissions in Europe and the costs and the potential to reduce VOC emissions. Similar work has been performed at IIASA on emissions of sulphur, nitrogen oxides and ammonia (Alcamo et al., 1990; Amann, 1989; Klaassen, 1991).
- Development of a simplified representation of complex models for tropospheric ozone formation, which can be incorporated into the RAINS model. The Norwegian Meteorological Institute has now an ozone model available (Simpson, 1991) which may be used for this purpose.
- Development of a software to incorporate VOC emissions and the ozone formation into the RAINS model and preparation of the users manual to facilitate the dissemination of the model.

This paper deals with the first task. Its objective is to present a concise overview of available emission inventories for VOC emissions in Europe, and data on costs and effectiveness of emission control technologies. Furthermore the paper suggests a design of the VOC emission and cost module of the RAINS-type of ozone model. The latter is clearly preliminary since the structure and input requirements determined by the atmospheric-chemical model yet to be developed are not fixed yet. Therefore, this paper should be regarded more as a discussion paper and as a quick reference rather than as a final design.

The structure of the paper is as follows. After briefly going into differences between the RAINS model and its conceived ozone counterpart (Section 2), Section 3 summarizes the currently available European emission inventories and the current developments in this area. Section 4 jumps at preliminary considerations with respect to the format of data relating to VOC emissions. The possibilities for constructing scenarios for the emissions of VOC are discussed in Section 5, while Section 6 explores the possibilities for creating a database on VOC control measures. The paper ends (Section 7) with conclusions and proposals for further work.

2 STRUCTURE OF A RAINS-LIKE OZONE MODEL

An overview of the data flow in the envisaged system is depicted in Figure 2.1. The model calculates "ozone-air pollution"¹, quite similar as "acid deposition" is assessed in RAINS. Starting point is a reference scenario or pathway for the developments of the emissions of ozone precursors (NO_x, VOC, CO) in the different European countries. Computation of the resulting "ozone-air pollution" - see below - is carried out with a module defining the relation between emissions and resulting air pollution. The latter is evaluated against desired levels of air quality. Next a set of control measures is proposed - an alternative emission scenario - and subsequently a new pattern of air pollution is calculated. A next evaluation, taking into account costs of control, may evolve into a new control scenario. Rules for choosing control measures may be such that, given a desired level of pollution, costs are minimal. In such cases cost-efficient control strategies evolve.

The feasibility of automation of such procedure depends on the type of relations between changes of emissions and resulting alteration of ozone concentrations. Practical application requires that such relations are linear:

¹ This "definition" has to be elaborated later in the development of the atmospheric-chemistry of the RAINS-like ozone model. It may obtain the form "frequency of exceeding ozone pollution standards" or in its integrated form, "excess ozone".

$$\delta O_3^{l,m} = \Sigma w_{i,j,k} * \delta E_{i,j,k}$$

where

- l,m,j,k = geographical (grid) coordinates
 i = type of ozone precursor, such as type of VOC
 $\delta O_{3l,m}$ = change in ozone concentration in grid l,m
 $w_{i,l,m,j,k}$ = weight factors (given a meteorology)
 $\delta E_{i,j,k}$ = change of the emission E of the precursor i (a VOC or NO_x) at location j,k.

The chemistry of atmospheric ozone formation is reputed for its non-linearity, therefore, an important research issue is to find those "windows" of conditions² - the magnitudes of deltas - where the equation is a sufficient approximation for carrying out calculations. There are indications that possibilities exist (Simpson, 1992), but further research is necessary. The approach taken in the construction of source-receptor matrices may have consequences for building the databases of both reference emissions and control measures. This will be treated in Section 3.1.

This briefly described methodology for calculations is quite similar to the approach in RAINS, differences being of practical nature. The difference relates to the size of the "window", or, in other words, the atmospheric-chemical conditions determining the viability of the assumption of linearity. In case of SO₂, NO_x, and NH₃ this window is quite large while in the case of ozone pollution its size will be much smaller. The practical implication might be that "strict emission control" scenarios cannot be evaluated against their environmental effects with only one source-receptor matrix. How to address this issue should be a subject for further research.

² An important parameter is the ratio between NO_x and VOC concentrations. At high or low ratios decreasing VOC respectively NO_x has hardly any effect on ozone concentrations.

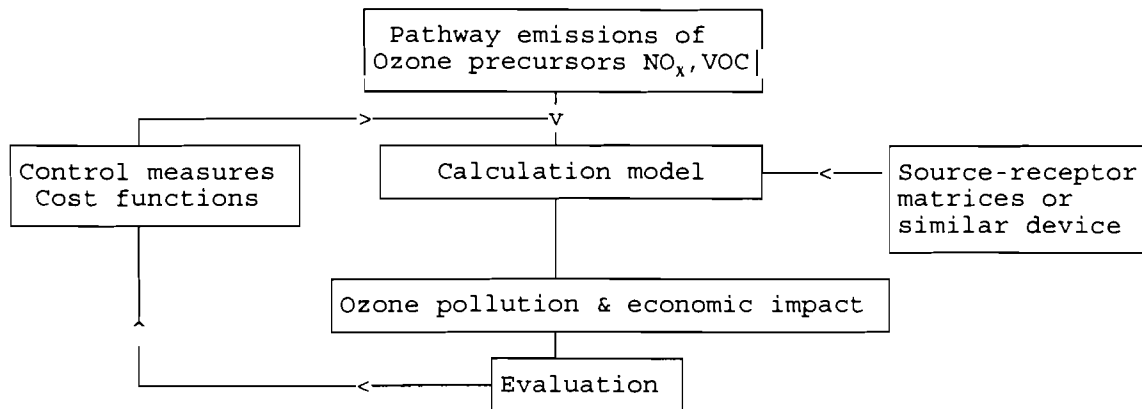


Figure 2.1. Data flows in the conceived model

Another complication, of economic nature, is due to the fact that in the ozone case control measures may affect emissions of different precursors simultaneously. A well-known example is the three-way catalyst for controlling tail-pipe emissions of cars. Such equipment reduces emissions of VOCs, NO_x and CO simultaneously. Other examples are measures to control emissions of solvents as solvents often are mixtures of VOCs.

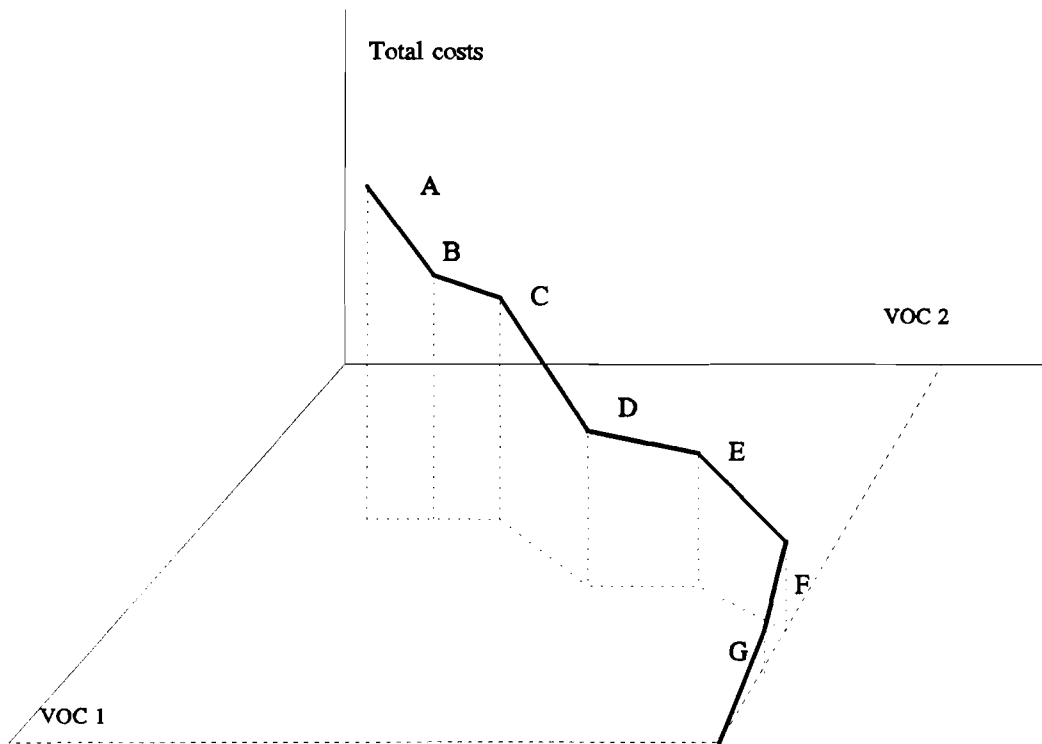


Figure 2.2 Cost curve for measures to control emissions of two ozone precursors.

Consequently the construction of abatement scenarios involves the use of joint cost curves, such as shown in Figure 2.2. This picture shows the costs of measures to control two ozone precursors: VOC_1 and VOC_2 (in reality cost curves will involve more types of VOCs and NO_x). The Figure shows emissions versus costs of measures A through G. Measures A, B and D operate only on VOC_2 , measures F and G only on VOC_1 , while measures C and E reduce emissions of both VOCs simultaneously. In fact, the term cost curve is misleading in this case, a more appropriate term would be cost surface, an n-dimensional cost curve (costs and n chemical species).

3 INVENTORIES OF EUROPEAN VOC EMISSIONS

3.1 Information sources

Inventories of European VOC emissions have been made within different institutional frameworks. Table 3.1 summarizes the characteristics of the inventories. The top six inventories are briefly described by Jourdan et al. (1990).

Table 3.1 Trans-boundary VOC-emission inventories in Europe

Program or initiator	Time span	Area	Remarks
TNO	1950-1980	13 countries	10 Western and 3 Eastern European countries. 4 types of stationary source categories.
OECD	1980	OECD countries	Comprehensive data set. Detailed source categories.
EEC Inventory	1982	EC countries	VOC speciation: non-reactive, unsaturated, others.
CORINAIR (EC)	1985	EC countries	Follow-up of preceding work. Development of methodology generally applicable. CORINAIR 1990 in progress. Chemical speciation limited to distinction of methane and NMVOC.
EMEP (UN-ECE)	1985	All Europe; 150*150 km grid	Data supplied by countries, if not own estimates (country totals are published by Jourdan et al. (1990).
PHOXA	1984	All Europe; approx. 25*25 km grid	Except for FRG and L own estimates. VOCs speciation: broken down to 10 model VOC (required by the photochemical model).
LOTOS (TNO)	1985	Eastern Europe (Veldt, 1991)	Western Europe covered by CORINAIR data. Includes a detailed chemical speciation.
EURAD-EMEP	1982	Eastern Europe (Pacyna, 1989)	Used in EMEP 1982 calculations.

The main differences between the inventories relate to:

- the spatial resolution (total area involved, grid size);
- the type of VOCs distinguished (Non-methane VOCs, other);
- the source categories involved;
- the source sectors distinguished.

The inventories have in common that the number of persons and institutes involved in these studies is very small (Baars et al., 1993). TNO in the Netherlands and NILU (Norway) appear to be the principal sources, in particular with regard to the inventories of Eastern Europe.

The focal point for large-area inventorying appears now to be the UN-ECE³. Recently (1992) a UN-ECE task force on emission inventories was established which, in its first meeting, recommended the use of the CORINAIR methodology⁴ as the preferred approach for all European countries (McInnes, 1992). The data of this inventory will be used in the EMEP program, including the long-term modelling of photochemical oxidants over Europe. CORINAIR concerns only stationary sources. The COPERT model, also developed for the EC, has a similar status in inventorying mobile sources.

The EMEP model distinguishes six characteristic man-made NMVOCs: ethane, ethene, propene, ethanol, n-butane and o-xylene. In a front-end model the inventory data are split to these VOC categories according to source-specific keys (Simpson 1991). The conceived atmospheric chemistry in the pursued ozone model will be derived from information created by these EMEP activities. The choice for using CORINAIR-derived emission data ensures thus an internal consistency in the ozone model.

3.2 A typical country inventory

Table 3.2 presents an inventory for the man-made emissions in the exemplary country Kakania, which is typical for most European countries. Road transport, gasoline distribution

³ Currently several institutions (OECD, UNEP, IPCC) are involved in world-scale emission inventorying.

⁴ Briefly summarized the CORINAIR methodology is a set of default emission factors (Bouscaren et al., 1992) - to be used in first order approximations - and a manual how to use them in inventorying. Data and calculation procedures are comprised in a computer program. This program has been distributed among environmental agencies in the various ECE countries. These agencies should gather emission data or, if these are not available, gather statistical data, which is according to CORINAIR relevant in emission estimating, and process these with the CORINAIR system. In the latter case default emission factors are used.

and refining (production of gasoline) together - these sectors can be regarded as mutually depending - account for the largest part of the emissions, closely followed by solvent use. Other sectors are distinctively less important. This table shows a typical pattern of emissions and the characteristic chemical speciation.

The table shows only anthropogenic emissions; emissions from natural sources vary widely over the countries, ranging from a few per cent of man-made emissions to over 100 per cent. Wood coverage (coniferous and deciduous) and, in particular, summer temperatures explain to a large extent the emissions and their temporal pattern.

Table 3.2 Man-made NMVOC emissions in Kakania

Source categories	Emission (share %)	Relatively main constituents in descending order
Road transport	48	
- gasoline vehicles	42	Alkanes, aromatics, alkenes
- other vehicles	4	Alkanes, alkenes, aromatics
- gasoline distribution	2	Alkanes
Refineries (non combustion)	3	Alkanes
Chemical industry (non combustion)	3	Alkenes, halogenated compounds, aromatics
Solvent use (not in chemical industry)	37	
- industrial	22	Alkanes, aromatics, oxygenated species
- domestic	15	idem
Stationary combustion	4	Alkanes, alkenes
Other	7	Fermentation processes: ethanol, emissions from coking: 50% alkenes

3.3 Uncertainty

The reliability of emission data decreases the more estimates are based on anecdotal data and "engineering judgment" and the less actual measurements are involved. Estimations of VOC emissions are particularly known for being based on non-measured information. Even in a country where statistical and marketing data are relatively abundant, like the UK, uncertainties are estimated to range from 25% to 100%, depending on the sector (Richardson et al., 1991, see also Baars et al., 1993, and Battye et al, 1987). The uncertainty is of particular significance in case of emissions due to the solvents use.

Lack of appropriate statistics and emission factors in Eastern European countries explains that in using Eastern European inventories typically one has to accommodate 100-200 per cent uncertainties. Moreover, in countries where large efforts have been made to assess emissions (California), ambient VOC concentrations cannot be properly reconciled with emission inventories (Fujita et al., 1992; Croes et al., 1993).

How to deal with uncertainties of such magnitudes will have to be decided from the results of sensitivity analysis with the dispersion models. An obvious research issue is to assess to what extent the source-receptor matrices to develop depend on the emission inventory.

4 FORMAT OF EMISSION DATA

4.1 Introduction

A key factor in the development of the model is the format of the data to be processed and produced in the model. With respect to emission data the issue is to what extent should emission data be detailed to:

- emission sources (sectoral format);
- types of VOCs (chemical speciation);
- information on costs of control measures;
- geographical characteristics of sources (spatial speciation and source characteristics

like stack heights).

For practical reasons it is not possible to use all available data, e.g., for each individual boiler. The issue is how to lump data, while preserving the essential information.

4.2 Data structure of required emission data

The structure of the database containing the emission data for a reference year is determined by the following constraints:

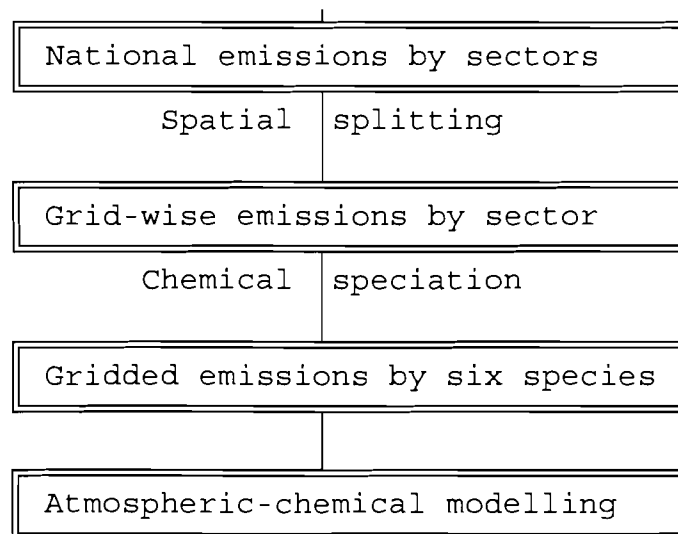
- the structure should be compatible with the structure of authorized data which are used in the international negotiations;
- the structure should be compatible with the structure of the databases on other photochemical precursors, in particular NO_x ;
- the structure should preferably allow the use of other (non-authorized) emission data which are expected to improve the reliability⁵;
- scenario development should be possible (emissions have to be tagged to relevant indicators (activities or emission explanatory variables), such as petrol consumption in case of emissions by cars);
- the structure should be adjusted to format of source-receptor matrices. The main requirements are that the data might have to be split into type of VOC, into location (spatial resolution) and temporal emission rates (diurnal cycles). To what extent these resolutions are necessary has to be judged from sensitivity analyses;
- data structures should be adjusted to format of abatement measures. The required database of abatement measures should contain the elements "effectiveness" and "costs". Eventual optimization of abatement strategies may depend on type of VOCs abated. If so, the type of VOC should be included (this is the case in policies to address short-term ozone problems).

Finally, it is necessary to take into consideration how the atmospheric model will be composed. Currently it is planned to construct these calculation devices with the long period

⁵ Good-quality anecdotal data of single plants or processes are sometimes found.

oxidant model of EMEP-MS-C-W (EMEP: the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe, MSC-W; Meteorological Synthesising Centre West, Oslo, Norway). This model describes the relations between the emissions of both VOCs and NO_x over Europe and the local occurrence of long-range transported photochemical oxidants, in particular ozone. Figure 4.1 shows how emission data are treated in the model (Simpson, 1991). Starting point are national emission inventories, made available through the UN-ECE. Currently these are national data obtained via UN-ECE; they are gridded by EMEP. The next step might be a division of the aggregate VOC emission data in typical VOC species: ethane, ethene, propene, ethanol, n-butane and o-xylene. It might be necessary to distinguish groups of VOC species depending on their role in O_3 formation.

Figure 4.1 Flow of data from European emission inventories to calculation model in the EMEP MSC-W long period oxidant model.



It is important to note that the model will be calibrated by comparing calculation results with measured ozone concentrations. Consequently, values of model parameters will depend on the emission data or, in other words, on the inventory. The same will hold for the functional relationship to be derived with the model. The conclusion with regard to the design of a RAINS-like model is to recommend the use of the same reference emission inventory as used in the calculation of the source-receptor matrices.

Since in the future the CORINAIR/COPERT inventory will be the European standard and because it is also used by the EMEP model the conclusion is to aim at using CORINAIR data in the model. However, the CORINAIR data relate to totals of emitted NMVOCs; a detailed chemical speciation is absent. Since the splitting of emission data is performed within the O₃ formation models, obviously their procedure should be copied in the RAINS-like model.

Sectoral format

The sectoral format proposed is derived from the CORINAIR format (Table 4.1). The main reason for this choice is that the emission inventory will be derived from a CORINAIR-structured database of UN-ECE. The table shows sectors on a two-digit level only, although the CORINAIR structure comprises also a third level.

Table 4.1 Major CORINAIR source categories (SNAP*) and selected activities

Source categories	Activities
Public power, cogeneration and district heating plants	Fuel consumption by both fuel and type of equipment
Commercial, institutional and residential combustion plants	Fuel consumption by both fuel and type of equipment
Industrial combustion in plants and processes with combustion	Fuel consumption by both fuel and type of equipment
Non-combustion processes	Oil refining (petroleum industry) (excluding combustion) Production of organic chemicals Wine making, brewing, bread baking (fermentation processes) Other
Extraction and distribution of fossil fuels	Storage and distribution of gasoline (including refuelling of cars) Offshore oil and gas industry
Solvent use	Paint and other VOC containing surface coating material Solvent for degreasing purposes Solvent use in manufacturing and processing of chemical products Other
Road transport (fuelling cars included in item 5)	Driving Gasoline evaporation from vehicles
Other transport	Not to account for
Waste treatment and disposal	
Agriculture	
Nature	

*SNAP: Selected Nomenclature for Air Pollution

The VOC inventory format, as proposed for the ozone model, consists of aggregate CORINAIR sectors. The sectoral format, shown by Table 4.2, is proposed considering the following:

- not too detailed for practical reasons, focusing on the main VOC-emitting activities;
- activity levels consist of relevant independent variables explaining the level of emissions;
- reflects the main target groups for control measures (road traffic, gasoline

distribution, solvent use⁶).

This proposal is preliminary. It might still be too detailed with respect to the uncertainty of data of emissions, scenarios, control measures and spatial and chemical resolution. An alternative, if the necessary level of detail cannot be proved, could be to restrict to two sectors, a "gasoline sector" and a sector "other". A decision about such a radical simplification can be made later when more information is available and after carrying out sensitivity analysis.

The source categories in Table 4.2 comprise two-digit level SNAP categories. The exact relation between the CORINAIR sectoral format and the proposed format is given in Section 5.

Table 4.2 A preliminary proposal for a sectoral format VOC emission database

Source categories	CORINAIR90 SNAP Code	Description
Road transport - gasoline vehicles - other vehicles	07, 09	Road transport, other transport
Gasoline distribution	0505	Gasoline distribution
Refineries (non combustion)	0401	Processes in petroleum industry
Chemical industry	0405 0603	Processes in organic chemical industry (bulk) Chemical products manufacturing or processing
Solvent use (not in chemical industry) - industrial - domestic	06 060408 060104	Solvent use, except for 0603, 060408 and 060104 (see below) Paint application: domestic use Domestic solvent use (other than paint application)
Stationary combustion	01, 02, 03	
Other	Other	
Nature	11	

¹ SNAP: Selected Nomenclature for Air Pollution

⁶ For instance, the EC is in the process of proposing directives directed at controlling emissions from gasoline distribution and from solvent use. EC directives setting emission standards for mobile sources are already valid.

4.2.2 Spatial format

The initial spatial resolution will be by country (regions), depending on the results of photochemical modelling. An important key for spatial distribution of national data grid resolution is population density (relevant for road traffic and small scale solvent use), whereas the location of large "point" sources like refineries, car or appliances manufacturing can probably be provided by the CORINAIR inventory and similar inventories for non-EC countries. The smaller the grid size the larger efforts will be necessary in spatial splitting of emission data. With respect to the spatial resolution it is also required to maintain consistency with the inventory used in the EMEP models.

4.2.3 Chemical format

Ozone is formed from various chemical substances through complicated photochemical processes. The organic compounds involved are collectively defined as VOC, although the photochemical activities of the species differ greatly, given all other conditions constant. However, "photochemical activity" is an ambiguous concept as some VOCs react fast in comparison with their rate of atmospheric dispersion and, therefore, add only to local ozone problems, while other VOCs react only slowly and do contribute to ozone formation over larger areas.

The concept of the photochemical oxidant creation potential (POCP) (Derwent & Jenkins, 1991), expresses the ozone forming ability (chemical reaction rates) of any VOC species relative to that of ethene. The background of the concept of the POCP is the desire to obtain guidelines for cost-effective control measures, as the cost-effectiveness of a control measure depends on its contribution to reducing ambient pollutant concentration on the one hand and on the costs involved on the other hand.

Table 4.4 Grouping of NMVOCs according to their POCP (in episodic ozone formation) (McInnes, 1992).

<p>More important</p> <p>Alkenes</p> <p>Aromatics</p> <p>Alkanes (> C6) except 2,3 dimethylpentane</p> <p>Aldehydes except benzaldehyde</p> <p>Biogenics (isoprene)</p> <p>Less important</p> <p>Alkanes C3-C5 alkanes and 2,3 dimethylpentane</p> <p>Ketones (MEK and Me-t-butylketone)</p> <p>Alcohol including ethanol</p> <p>Esters except methylacetate</p> <p>Least important</p> <p>Methane, acetylene, benzene, benzaldehyde, ketones, methanol, methylacetate and chlorinated hydrocarbons.</p>
--

In the current version of the EMEP model, however, only six typical species (see Section 4.2) are distinguished. The use of the POCP concept seems not appropriate in addressing typical (long range) trans-boundary ozone pollution (Simpson, 1991). Above it was already concluded to stick close to the EMEP procedure for speciation to preserve consistency between the emission data and the ozone formation model yet to be developed.

5 VOC PATHWAYS - BUILDING A REFERENCE SCENARIO

The minimal requirements for construction of VOC emission scenarios are

- the availability of a forecast of the development of an activity and
- an emission factor related to this activity (emission explanatory variable).

This emission factor should preferably account for autonomous technological developments (e.g., developments influencing emissions though not prompted by new environmental policy

measures). The procedure can be simply expressed as:

$$E_k^i(t) = A_k^i(t) * [T_k^i(t) * E_k^i(t_0)] \quad (i \text{ for country/region } i)$$

where

- $E_k^i(t)$: annual emission in the year t from source k
- $A_k^i(t)$: value of emission explanatory variable (such as fuel consumption, consumption of feedstock, production value of a sector, traffic performance)
- $T_k^i(t)$: factor representing autonomous technical development influencing emissions (accommodates factors as renewal of capital stock and influence of policy other than in environmental areas, such as industrial health and safety)
- $E_k^i(t_0)$: emission factor in reference year

This section summarizes, sector by sector, the available information about trends in activity levels or, if not available, the possibilities to develop this kind of information, followed by a similar discussion on emission factors.

5.1 Road transport

Road transport is the most important sector generating VOC emissions; in Western Europe cars do account for about half of the total anthropogenic VOC emission. The share of road traffic in man-made VOC was estimated at 4.7 Mt of a total of 11.3 Mt anthropogenic VOCs for all European OECD countries in 1980 (OECD, 1990).

As in the NO_x case, transport is the main source of emissions. However, whereas NO_x emissions are due to the use of both diesel and gasoline engines, VOC emissions mainly result from the use of gasoline as an automotive fuel. Another difference in respect of NO_x emissions is that two-stroke engines, used in mopeds, motorcycles and in Eastern European (former GDR, Hungary) cars, do emit large amounts of VOCs in comparison with NO_x . A further difference is that, while NO_x is only a tail-pipe pollutant, VOC is released via other sources - evaporating from petrol tank and fuel system - as well. Emissions due to

"breathing" of gasoline tanks should be related to the number of cars and not to mileage.

A sophisticated approach to forecast national road-transport emissions (Klooster, 1987) starts with an economic scenario of the future per-capita consumption and estimates car trips by various parameters such as mean speed, mileage per trip and type of car (rated power, type of motor, emission abatement). By multiplication with the appropriate emission factors⁷ future emissions can be derived. Autonomous technological development should embody existing environmental legislation concerning car emissions.

This approach requires an extensive amount of data, ranging from econometrically derived price elasticities of fuel consumption and car purchase to engineering data of abatement measures. The short-cut alternative is to use aggregate emission factors which relate emissions to fuel consumption, as it has been done in the current RAINS model for NO_x emissions. The main advantage of this method in scenario building is a practical one; scenarios of fuel use are readily available⁸ whereas scenarios for car mileage are only available for a small number of countries, if any. The disadvantage of the aggregate method is its limited accuracy. However, a solution of the latter problem is offered by the COPERT system. COPERT is a computerized procedure for calculation of annual emissions of car fleets taking into account all technical factors mentioned above. It calculates also fuel consumption and, thus, emissions by fuel use. COPERT is developed as a tool for inventorying transport emissions. Recently, a model (FOREMOVE) (Samaras et al., 1991) to estimate future emissions has been constructed based on the COPERT database. This model calculates future emissions from estimated future vehicle fleets, taking into account annual rates of renewal, the increase of the stock of vehicles, and the effects of exhaust gas regulations. This procedure is not compatible with a fuel-use based prediction procedure⁹. However, a similar approach as in the FOREMOVE system might be used, by calculating

⁷ Relating emissions to distances (g/km).

⁸ They are usually developed as a part of energy scenarios.

⁹ It would if fuel use was calculated from the extent, composition and use of future vehicle fleets in the various countries. However, development of FOREMOVE and UNECE energy-scenario compiling are not explicitly coordinated.

vehicle use (mean mileage, mean speeds) data from scenarios for fuel use and for vehicle fleet, and subsequently adding information on car-emission abatement. Obviously, neglecting the relation between exhaust-emission abatement and fuel consumption introduces an error. However, it can be argued that its magnitude will be small compared with other uncertainties.

A characteristic of the reference scenario for transport is that technological change due to legislation has to be taken into account (rate of renewal of the car fleet). Most countries have adopted the same (ECE/EC) legislation with respect to car emission. It may be expected that in due time all cars will meet ECE and EC standards. Another technological change is "autonomous", the phasing out of obsolete type of cars and trucks in Eastern Europe. The production of cars using two-stroke engines has been terminated and it is expected that these type of cars will disappear eventually. The same holds for petrol-engine driven trucks.

Gasoline engines

It is concluded that a reference scenario for VOC emissions from mobile sources (gasoline) can be built in a similar way as currently NO_x emissions are dealt with in the RAINS model, i.e., based on fuel consumption. VOC emissions due to evaporation from gasoline tanks, however, should be calculated from car fleets (number of cars).

Other transport: Diesel engines

The second type of road transport comprises vehicles equipped with diesel engines. VOC emissions of diesel vehicles are typically less (a quarter to a third) than those of vehicles equipped with gasoline engines. A scenario can be built along the same procedure as described above for gasoline vehicles.

VOC emissions from diesel engines used in inland shipping and rail transport are low (emission factor estimated at 2.4 g/kg diesel fuel (Lloyd's Register, 1991)).

5.2 Gasoline distribution

Gasoline distribution is a two-digit subsector of the CORINAIR sector "extraction and

distribution of fossil fuels". The emissions are closely related with distribution of gasoline. Therefore, gasoline consumption is an appropriate "rate of activity" to be used as an independent variable for developing a reference scenario. Typical problems, however, are caused by large differences in the equipment used in the different countries. These differences relate to:

- type of storage tanks (fixed roof versus floating roof);
- practice of loading (splash loading versus bottom loading),
- the use of equipment to prevent losses (or emissions) of gasoline, such as vapour balancing equipment;
- the number of transshipment of gasoline along its way from refinery to gasoline station.

Aggregated information (on a two-digit level) will be available with the forthcoming CORINAIR90 inventory. A comparison with gasoline consumption data will reveal the technical differences among countries.

5.3 Refineries - non-combustion

Refineries are also contained as a two-digit subsector in the CORINAIR SNAP nomenclature (subsector of non-combustion processes). The handling and storage of the volatile products constitute the main origin of the emissions. Rates of activity, which explain the development of emissions, might be the amount of crude oil processed - currently used in CORINAIR - or the amount of gasoline (and naphtha) produced. Energy scenarios have both types of information available.

Country-specific state-of-the-art of refining (influencing the overall emission factor) differs greatly among countries. In CORINAIR the default emission factor for refineries is 0.5 kg per tonne crude processed. This figure is an estimate for Western European countries; estimates for Eastern European countries, such as refineries in CIS countries, are up to a factor of twenty higher (Veldt, 1991). These differences can be explained by the lack in technical development and investments over the years, although in Eastern Europe on the other hand the share of light products - the volatile ones - is lower than in typical Western

European refineries, which usually have hydrocracking capacity for producing light products from heavier products.

While for Western European countries scenarios for VOC emissions can be based on the expected development of crude oil throughput, in the case of Eastern European countries it appears necessary to take into account the rate of renewal of the refinery industry.

Table 5.1 Emission factors (kg/tonne) for refining

Country/source	
(West) Germany	Trend in emission factors: 1966-70: 2 kg/tonne; 1974: 1.5; 1978: 1.0; 1982-1986: 0.4 (Veldt, 1991)
Concawe refinery	0.4 (typical Western European refinery) (Foster et al., 1987)
CORINAIR refinery	0.8 (Bouscaren, 1991)
USA	1980: 0.65 (Veldt, 1991)
Poland	1989: 0.88 (Plock refinery) (Veldt, 1991)
USSR	1985: 12 (refining losses (% of output) 1975: 1.51; 1980: 1.26 (Veldt, 1991)) 6 kg/tonne (Pacyna, 1989)

5.4 Solvent use

This sector comprises the two-digit CORINAIR sectors with the following exemptions:

- solvent use in chemical industry (see above)
- domestic solvent use: a three-digit level category in the CORINAIR system.

Industrial, excluding chemical industry

The main subsectors are:

- surface coating in the metal industry (cars, appliances, other);
- painting in the textile industry;
- painting in flatwood and wooden furniture industry;
- use of (hexane) solvent in the vegetable oil industry;
- architectural painting;

- solvent use for degreasing purposes.

These sectors have little in common except the use of paint and lacquer. It is obvious that there is no common denominator suited for use as an aggregate emission explanatory variable (rate of activity). Lacking alternatives, one might choose industrial development - measured as development of aggregate production values - as a surrogate for $A^i(t)$. Growth in production levels $A^i(t)$ will lead to increased emissions. However, considering the factor $T^i(t)$ on an equally aggregate level, one may argue that the trajectory of technological development is directed towards less use of solvents. Arguments are:

- environmental incentives in technologically leading countries;
- considerations of occupational health;
- safety reasons (fire protection and insurance costs).

Taking into account the considerable uncertainty in the emission figures (-50% to +100%; see Baars et al., 1993) it appears to make little sense to put extra efforts in scenario constructing. As a first estimate, assuming no net changes - as the result of an off-set from increase in production and technological development - seems to be a good approximation. Of course, in cases where elaborated scenarios are already available these should be adopted. This is for instance the case in the Netherlands (KWS2000) (scenarios up to the year 2000).

If atmospheric-chemical modelling would indicate which VOC are of major importance in trans-boundary pollution, a separate effort directed at such emissions might be justified.

Domestic solvent use

Scenario construction for this sector is hampered by similar problems as indicated above. Therefore, the suggested approach to arrive at a first estimate would be to keep these emissions constant, unless more specific relevant information is available. Table 5.2 presents the results of a recent inventory of VOC sales in the Netherlands.

Table 5.2 Main VOC-containing products in the Netherlands in 1990 (Pearce/KWS2000, 1992)

Product	Emissions tonnes	Per capita (kg/a)	Remarks
Paint and varnishes	30,000	2.1	
Thinners	7,160	0.52	Connection with the consumption of VOC-based paint (cleaning brushes).
Aerosol hairsprays	3,800	0.27	Sales expected to increase sharply. Atomizer alternatives marketed.
Stain removers	2,048	0.15	
Methylated spirits	1,647	0.12	Cleaning agent, but also used as ignition agent (no emission in the latter case!).
Aerosol deodorants	1,238	0.9	
Window cleaner	1,205	0.9	
Adhesives	600	0.4	13,000 tonnes institutional use.
Other	7,000	0.5	A very large variety of products.

The per-capita consumption of these products varies among the EC countries (Olsthoorn et al., in prep.).

5.5 Chemical industry

Chemical industry comprises the SNAP two-digit subsector "Processes in organic chemical industries (bulk production)", which is a subsector of "Non combustion processes" and the two-digit subsector "Chemical products manufacturing or processing", being a subsector of "Solvent use". The proposal to establish a sector "Chemical industry" deviating from the CORINAIR main categories is based on two arguments. The first is that the emission profile of chemical industry is characterised by the occurrence of emissions of VOC with a distinct photochemical character (see Table 5.3). In particular alkenes (ethene, propene) constitute a major share. The importance of the different emissions for trans-boundary pollution issues may, consequently, diverge widely.

Table 5.3 Chemical speciation of the VOC emission by the Dutch chemical industry in 1985 (Thomas & Olsthoorn, 1987).

Type of chemical	Emission (tonnes)
Aliphates, saturated	4,300
Aliphates, unsaturated (mainly ethene)	5,100
Aromatics	2,600
Halogenated hydrocarbons	6,100
Aliphatic hydroxyl compounds (alcohol type)	1,600
Other oxygenated hydrocarbons	2,510
Organic nitrogen compounds	450
Other	1,530
Total	24,400

The second argument is that processes in the chemical industry are to a large extent coupled or joint and, therefore, for reasons of consistency these should be treated in a coherent manner.

A procedure to develop scenarios taking into account the law of conservation of mass has been developed for the Dutch chemical industry (Olsthoorn, 1987). This procedure creates surveys on expectations of final demand for chemical products, and calculates subsequently production levels, making use of chemical mass balances (input-output approach). In such an approach the "rate of activity" is represented by a vector of product-specific production levels. Calculation of future emissions is performed by combining these production vectors with vectors of VOC emission factors (e.g. CORINAIR default factors). At the moment it is not clear whether this method will be feasible on a European scale. Considerable research efforts will be required and, for the time being, this seems not an appropriate method.

Allemand et al. (1990) created scenarios on emissions in the EC-wide chemical industry. Their method was based on a set of emission factors for chemical key products (key in the sense of VOC emissions). Scenarios were made by assuming some average annual growth

in the outputs of the products. A method for taking into account autonomous technological development was developed by comparing emission and production data from different countries.

A simple alternative to these elaborate methods would be to use production values of the chemical industries in the various countries as "rates of activities" and appropriate emission coefficients¹⁰. If a scenario for the economic development of the chemical industry is available, it will be possible to obtain an estimate of future emissions. The drawback of this method is the loss of relevant information for estimating autonomous technical development and autonomous change in industrial structure (shifts in the product mix) as well as for assessing impacts of control measures.

Considering the relatively small contribution of the chemical industry to VOC emissions, the large uncertainties in the emission data and the large effort needed to develop elaborated scenarios, it is recommended to use constant emission over time as a base scenario.

5.6 Combustion in stationary equipment

This category comprises the following three one-digit energy sectors of the CORINAIR system:

- Public power, cogeneration and district heating plants;
- Commercial, institutional and residential combustion plants and
- Industrial combustion in plants and processes with combustion.

Scenarios for fuel consumption can be derived from available energy scenarios.

5.7 Other sectors

Other sectors emitting VOC are fermentation processes, other transport, waste treatment and disposal, coking and agriculture. Fermentation processes involve baking of bread and

¹⁰ An emission factor relates emission to a rate of activity in physical terms, while a similar coefficient relates emissions to monetary values.

production of beer, wine and spirits. The VOC emitted is mainly ethanol. Demographic scenarios might be used to make a forecast of these emissions.

"Other transport" relates to inland shipping, shipping in coastal waters and at open sea, air transport. The development of these emissions can be related to energy consumption. "Waste treatment and disposal" and "agriculture" mainly emit methane and, therefore, are only minor sources of non-methane VOC which could, in a first approximation, be considered as constant over time.

5.8 Conclusions

This section treated, sector by sector, the possibilities for building reference scenarios to describe the emission development. In the various countries about half of the VOC emissions is related to consumption of fossil fuel, consequently half of the VOC emissions can be tagged to energy pathways in the current RAINS model. A problem to be resolved is the split in fuel use for transport into the required categories (petrol versus diesel, road versus other transport, vehicle fleet characteristics).

The trend in the development of the other half of the emission - mainly due to solvent use - appears as indistinct: a growth in emission-causing activities appears likely to be offset by technological development directed towards less use of solvents. Considering the large uncertainties in the emission estimates a reference scenario in which VOC emissions from solvents are kept constant will be sufficient as a first estimate.

Minor exceptions, with respect to the magnitude of the emissions, are the chemical industry and combustion in stationary equipment.

Table 5.4 Building VOC emission pathways: a summary of methodology and available data

Sector/process	Activity rate	Availability
Gasoline powered vehicles		
- Four-stroke engines	Gasoline consumption, rate of renewal	RAINS
- Two-stroke engines	Gasoline consumption, rate of renewal	RAINS
Diesel powered vehicles	Middle distillates	RAINS
Gasoline distribution	Gasoline consumption	RAINS
Refineries	Crude throughput	RAINS
Solvent use	Keep constant	
Chemical industry	Keep constant	
Combustion in stationary equipment	Fuel combustion	RAINS

6 ABATEMENT OF VOC EMISSIONS

One of the features of the RAINS model is its database of country-specific cost functions, relating country-aggregate marginal control costs with emissions of SO₂, NO_x and NH₃. A RAINS-like ozone model requires a similar database. However, two differences appear to exist. The first, of fundamental nature, is that due to the complicated chemistry, emission reductions will not necessarily result in proportional decreases in ambient ozone concentrations. An atmospheric-chemical analysis is needed to address this issue.

The second difference, of practical nature, is that the number of possible technical measures to control VOC emissions are an order of magnitude larger, corresponding to the larger number of different types of sources. In section 2 it has been discussed that control measures often address more than one ozone precursor; this constitutes a third difference.

This chapter briefly reviews the currently available information about both abatement of VOC emissions (add-on technology) and change in techniques (phasing out of the use of two-stroke engines in cars). The emphasis is on removal efficiency and cost-effectiveness. If possible

it is attempted to specify these data with respect to sector (sectoral format as proposed in table 3.2) and country.

6.1.1 Road transport - gasoline vehicles

Policy

The commonly used policy instrument to control air pollution from vehicles is setting of product standards. These standards usually address the tail-pipe emissions of CO, NO_x and VOCs simultaneously. Distinctions are made between gasoline cars and diesel cars. In the Western European countries the current standards require the use of a three-way catalytic¹¹ device for gasoline cars.

Gasoline cars

The current database of abatement measures of the RAINS model already contains descriptions of measures which reduce (tail-pipe) VOC emissions from petrol-powered cars: measures taken to comply with UN-ECE standards. The well known example is the use of a three-way catalyst device which simultaneously removes VOC, NO_x and CO from the exhaust gases. In EC countries all new cars sold are required to be equipped with such a control device (four-stroke spark-ignition engines only).

Table 6.1 presents provisional estimates of aggregate emission factors (emissions related to gasoline consumption) and summarizes possibilities for emission control and estimated mean costs.

¹¹ Controlled loop systems, where the air-fuel mixture is controlled in order to stoichiometric combustion conditions in the engine (Allemand et al., 1990).

Table 6.1 Gasoline passenger cars in EC countries, aggregate tailpipe emissions, their control and (capital) costs involved

Technology/standards	NO _x g/kg	VOC g/kg	Average extra purchasing costs US\$/vehicle	
Base case (1980)	24	18	-	
EC-15-04	7	7	400	Large cars catalysts, medium and small cars equal standards
US-83 standards	7	2	800	All cars catalysts. Adopted by EC.

Derived from Allemand et al. (1990). Costs to customer before tax. Costs and benefits of changing fuel consumption and maintenance costs ignored. Equivalent data are presented in the RAINS model.

VOC emissions from gasoline cars in Eastern Europe are estimated to be double the Western European 1980 emissions due to the use of two-stroke engines in cars (Veldt (1991): 32 g/kg, Pacyna (1989) 37 g/kg (includes evaporation emissions)).

An extra measure, specifically addressing only VOC emissions, is the use of a carbon canister to prevent emission of gasoline vapour from gasoline tanks and from the fuel system (such as the carburetor - hot soak emissions). These emissions are estimated at 7 g/kg fuel (Veldt, 1991). A small canister (1 litre) is sufficient to absorb VOC released by hot soaks and by diurnal temperature cycling (breathing of tanks), a large one (6-8 litres) can absorb VOC released at refuelling of car tanks (Stage 2 policy, see section 6.2). Accommodating both control measures requires data of the number of cars. Data of cost effectiveness are presented in table 6.2.

Table 6.2 Controlling evaporative and refuelling emissions with carbon canisters, effectiveness and costs (Derived from CONCAWE 6/87, 1987)

Type of emission	Type of control	Efficiency	Costs	
			US\$/tonne abated	US\$/vehicle
Evaporative losses	1 litre canister	90%	300	20
Refuelling losses	8 litre canister (additional)	90%	1350	100

Average life of equipment 10 year, maintenance costs ignored. Costs of control of refuelling emissions are **additional** to control costs of evaporative emissions. (CONCAWE 6/87). Total annual costs. Reference emissions (kg/tonne fuel).

6.1.2 Road transport - other vehicles

VOC emissions of diesel engines may be reduced by application of (trap) oxidation catalysts. These devices, however, are meant to reduce emissions of suspended particles. Currently in some countries experiments are carried out (Sweden, The Netherlands, FRG), mainly for buses and lorries. The use of catalysts, however, requires the use of low-sulphur diesel the production of which involves higher costs. Theoretically, oxidation will also affect VOCs in diesel exhaust gases, but the magnitude of this effect is not known yet.

6.2 Gasoline distribution

Evaporation of gasoline is a source for VOC air pollution which is addressed in the USA and in Western Europe by a specific policy¹² aimed at controlling the emissions along the chain of gasoline distribution from refinery dispatch to the refuelling of car tanks (Figure 6.1). Displacement emissions constitute the main category of emissions, they are abated by vapour balancing systems and vapour recovery units (VRU). Vapour balancing is appropriate when tanks with fixed containments (fixed-roof storage tanks, car tanks) are loaded from similar tanks: the gasoline vapour displaced from a receiving tank is returned to the tank from which

¹² The CEC is preparing a specific directive addressing these emissions.

the gasoline is delivered (venting into the atmosphere is thus prevented). At the front of the system the vapour cannot be received (see Figure 6.1); the solution to prevent emissions is usually to recover the vapour with a VRU (or incineration).

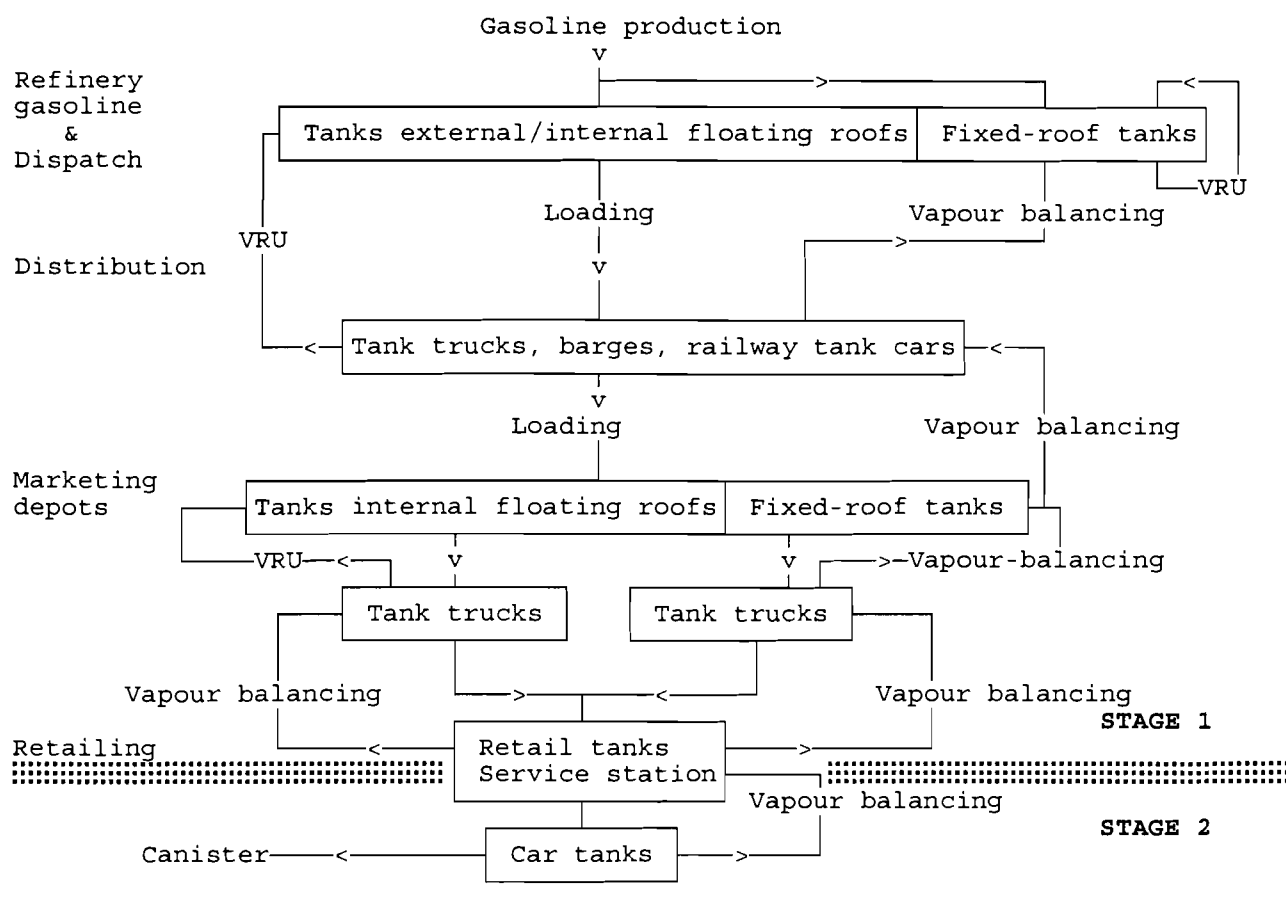


Figure 6.1 Gasoline logistics. From production to car (see text)

Figure 6.1 displays the characteristic elements of the gasoline distribution system. Currently, the use of fixed-roof tanks in gasoline storage is not common in Western European countries. Usually two groups of control measures are distinguished: Stage 1 and Stage 2¹³ measures. Stage 1 measures refer to emission control along the chain from refinery to delivery of fuel at the service station. Vapour collecting at the service stations is referred to as a Stage 1B measure, whereas the other Stage 1 measures (vapour balancing only at loading of trucks combined with VRU) are referred to as Stage 1A measures. Stage 2 refers to control of the

¹³ The origin of this terminology is the discussion in the USA around the introduction of these measures.

emissions at the filling of car tanks.

Table 6.3 A brief overview of emission sources, emission factors, control measures and cost effectiveness (from Richards et al., 1990). See text for explanation of cost figures.

Source	Emission factor (% on throughput)	Control measure ³ (% effectiveness)	Cost effectiveness (US\$/tonne)
Terminal tankage			
- fixed roof tanks	0.19 ¹	Internal floating roof (90-95%)	1000 (see Table 6.5)
- floating roof tanks	0.02		
- internal floating roof tank	0.009		
Truck loading			
- top splash	0.105	Change to submerged loading by modifying Vapour Recovery Unit	
- top submerged	0.055	Vapour Recovery Unit (80%)	
- bottom	0.050	Vapour Recovery Unit (90%)	
Rail car loading			
- top	0.06	Vapour Recovery Unit (80%)	
Ships/barge loading			
- bottom	0.04	Vapour Recovery Unit (80%)	not clear
Service stations			
- displacement	0.14 ²	Additional vapour bal. (95%)	1300-9000 (single stage ⁴). This figure relates to costs of both the vapour collection and recovery (see above).
- other origins	0.02	(Various, good housekeeping)	
Car-tank filling			
- displacement	0.18	Vapour balancing (56-86%) ⁵ Large on-board canister (95%)	5820 (in case of 56% average effectiveness)

¹ Mainly displacement emissions at loading.

² In case of a gasoline with a true vapour pressure (TVP) of 35 kPa, which is typical for Western Europe. Vapour balancing at this point in the distribution system requires, of course, recovery of the collected vapour in the front end of the system.

³ Only the main control measures. Among other measures are "Improved seals", "Good housekeeping".

⁴ Double-stage VRUs have recovery rate 99.99%, while this figure is 96% in case of single-stage VRUs.

⁵ 56% refers to average practice in Switzerland and Sweden (1988), while 86% refers to the situation in the USA (Richards et al., 1990).

Gasoline distribution. Stage 1

These measures are dealing with emissions due to loading of tanks, tank trucks, barges and car tanks and with breathing emissions of tanks. Table 6.3 summarizes the various types of emissions and technical possibilities for control. The reference cases indicated in the table represent the state-of-the-art around 1970.

The economies-of-scale are important in assessing the cost effectiveness of measures as both the capacity and annual throughput determine the costs and also the required recovery rate of the VRU used: the more recovered the higher the costs¹⁴. This explains the cost range. According to Richards et al. (1990) costs increase sharply from the 2000-3000 US\$/tonne range when depot throughput falls below 300,000 tonne/yr. From the data given by Richards et al. (1990) it is estimated that the cost effectiveness of Stage 1A measures are about 50% less than Stage 1A/B measures.

Gasoline distribution. Stage 2

Stage 2 control refers to the recovery of gasoline vapours released at filling of car tanks. The effectiveness of the vapour balancing systems varies in practice (USA, Sweden, Switzerland) between 53% and 86% depending on the level of regulatory enforcement (Richards et al., 1990).

The alternative is on-board (carbon-canister) vapour recovery, as discussed in Section 6.1. Note that the effectiveness of the on-board canister is estimated by the refinery industry (CONCAWE) at 95% (Richards et al., 1990), while other sources (Allemand et al., 1990) mention 80% effectiveness.

Gasoline quality

Evaporative losses of gasoline are a direct function of the volatility of the gasoline. This fuel property is often indicated as the RVP (Reid Vapour Pressure). The fuel RVP is adjusted at the refinery along with the type of climate and season, in winter high, in the summer low.

¹⁴ The common techniques are the use of absorption equipment or cooling of vapours in order to condense the vapour. The deeper the cooling - more powerful cooling equipment - the more gasoline is recovered. Uijlenbroek (1991) reviews currently available techniques.

Adjusting gasoline RVP to environmental requirements appears to be very costly compared with the other measures (Amadei, 1992).

Conclusions

How to abate evaporative emissions due to the use of gasoline is well known, however, only at an engineering level. In order to estimate cost-curves for countries information is needed about the structure of the gasoline-supply system.

6.3 Refineries

Table 6.5 gives an overview and classification of (non-combustion) refinery emissions, while table 6.6 attempts to summarize some of the data about cost effectiveness of abatement measures.

Table 6.5 Classification of refinery VOC emissions

Category
PROCESS
Leaks through valves, flanges, compressors and pumps (fugitive/diffuse emissions)
Periodical shut down of process units (maintenance procedures)
Catalysts regeneration
Waste water treatment (in particular oil-water separation and settling ponds)
Safety systems (flares and pressure reliefs)
STORAGE OF FEEDSTOCK AND PRODUCTS
Breathing
Leaking through seals of floating roofs
LOADING
Drive out of vapour/working emissions
Through seals

Table 6.6 Summary of literature data on control measures, costs and other aspects.

Description.	Cost effectiveness	Source	Remarks
ABATEMENT OF PROCESS EMISSION			
Inspec. & maintenance valves & flanges	- 0.22 US\$/kg abated	Radian as cited in Jourdan (1990, p.279)	This figure is a function of the value of distillates retained and of the reference leak rate
Inspec. & maintenance in a typical refinery	Dfl 5/kg abated ECU 0/kg abated	Annual costs (KWS-2000, 1989) Effectivity 60% (fugitive emissions) Radian as cited by Jourdan (1990), p. 279	
Inspec. & maintenance pumps and compressors	- 91,000 US\$/a	No. of valves 13.300. Radian as cited in Jourdan (1990).	
Collecting vapours or liquids escaped from safety devices, piping and recovery or flaring.	2-4 US\$/kg abated		
Recovery of evaporated VOCs by enclosure of waste water treatment equipment and recovery or flaring.	Unknown		
Floating covers	Benefit 0.5 US\$/kg abated ECU 0.13/kg abated	EPA, 1978 Concawe 1987. Annual costs. Effectiveness 90%	
IMPROVING STORAGE OF FEEDSTOCKS AND PRODUCTS			
Internal floating roof (retrofit or new) in fixed roof tanks containing gasoline and the like	Benefit (Return of inv. 1 2 year) 0.13 kg/ton gasoline abated Dfl 2/kg abated	Jourdan (1990) p. 290). Magnitude. Depending on RVP. Annual costs. Specific investment Dfl 20/kg (KWS-2000, 1989).	
Improving seals of floating roof tanks	Dfl 0.4/kg abated ECU 0.8/kg abated	Annual costs. Investment Dfl 8.5/kg abated (KWS-2000, 1989). Annual costs. Effectiveness 83% (incl. inv. costs) (Concawe, as cited by Allemand, 1990, p.107 & 133)	
KWS-2000: It is not clear whether annual costs include the benefits of emission reductions. The same holds for capital costs. Costs given by Radian depend on the market value of hydrocarbons (naphtha, gasoline). ECU data given by Allemand Vol 2 (1990), p. 132. These data are derived from a Concawe study. 1 ECU = 1 \$ = 2.3 Dfl			

Table 6.6 shows that in this sector several control measures may bring about economic benefits or are at least cost-neutral. In particular in Eastern European countries the refining sector is technologically at a low level compared with North-West European standards. For example, 70% of USSR refinery equipment is characterized to be obsolete (O&G J, Feb. 12, 1992). Consequently, the data from Table 6.6 show that the costs of improving the environmental characteristics of the Eastern European refinery sector are low or even negative.

A comprehensive overview of emissions, possibilities for abatement and costs of abatement has been made for a hypothetical refinery by CONCAWE (Foster et al., 1987). Results of this study are summarized in Table 6.7, while Figure 6.2 presents these results graphically.

This refinery is an average Western European one, having hydrocracking capacity. This contrasts with the average Eastern European refineries. One may argue that the production of light - volatile - products is less in these refineries. This might partly offset the effect of large fugitive emissions.

Table 6.7 Emissions and measures for emission abatement in a hypothetical refinery (capacity 5 million tonnes annually (Foster et al., 1987))

Emission source (abatement)	Emission (tonnes/yr)	Abated (tonnes/yr)	Cost effectiveness (US\$/tonne)
Fugitive losses (I&M)	1300	750	100
Waste water treatment (FC)	1120	180	460
Condensate tankage (RSS)	1102	18	600
Light naphta (RSS)	1091	11	650
LCN tankage (RSS)	1080	11	850
Gasoline tankage (RSS)	1048	32	1030
Poly gasoline tankage (RSS)	1042	6	1160
Road (VRU)	889	153	1560
Crude tankage (RSS)	816	73	1650
Reformate tankage (RSS)	810	6	1920
HCN tankage (RSS)	807	3	2800
Naphta tankage (RSS)	774	33	2840
Rail (VRU)	722	52	3760
Reformer feed tankage (IFC)	716	6	4930
Ship/ocean barge (VRU)	643	73	6450
Barge (inland) (VRU)	599	44	8510
Residue tankage (IFC)	593	6	11145

I&M Inspection and maintenance, IFC Internal floating cover (in fixed-roof tanks), FC Floating cover (external), RSS Rim-mounted secondary seals (of floating roofs of storage tanks), VRU Vapour recovery unit.

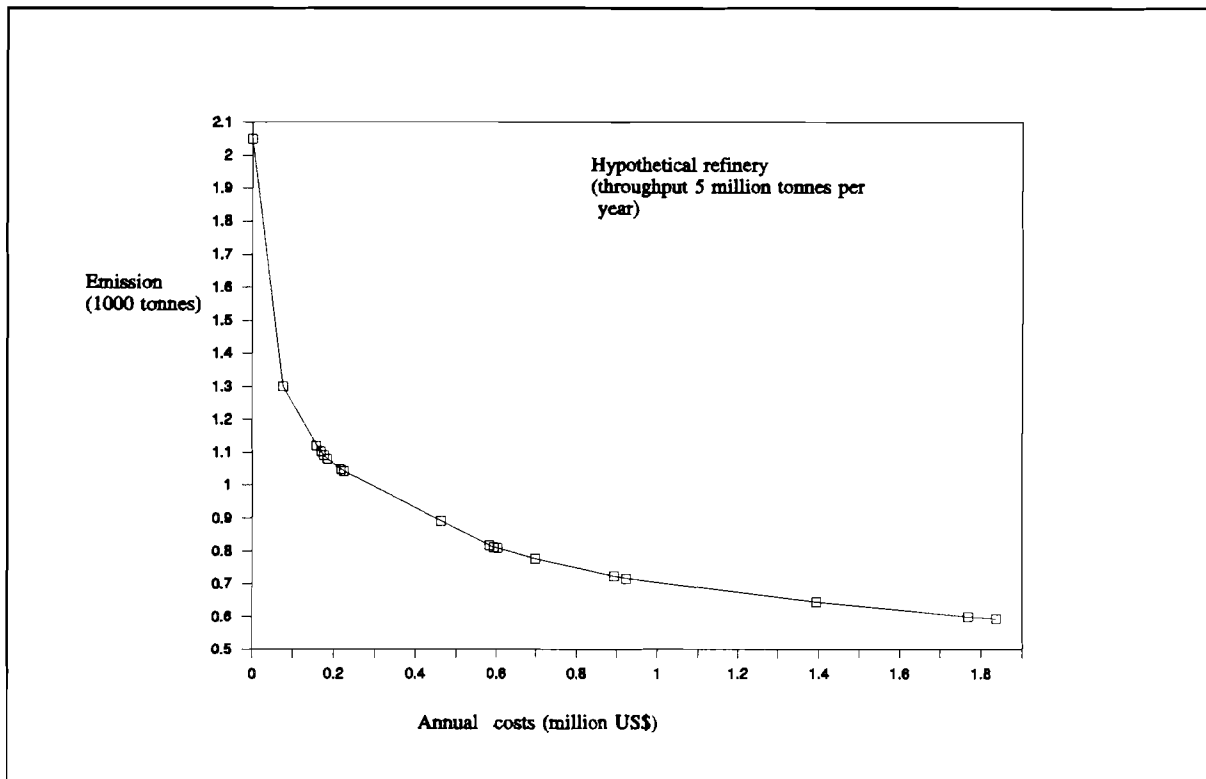


Figure 6.2 Emissions and annual abatement costs of a refinery (Foster et al., 1987). Excluding emissions from ballasting crude oil tankers.

6.4 Chemical industry

The chemical industry is, in a technological sense, an extremely heterogeneous sector and possibilities and costs of control of VOC emissions will diverge widely. A sense of possibilities for control might be gained through a statistical analysis from the characteristics of the sources of emissions. Such statistics, although crude, may be derived from Table 6.7 and other literature. The result might be a generalized cost curve (Figure 6.3).

Table 6.7 Source characteristics, abatement measures, their effectiveness and costs, in the Dutch chemical industry*, as expected for the year 2000 (KWS2000, 1989).

Source type	Emission kt/y	Control measure	Reduction kt/y	Investment MUS\$	Operating costs/MUS\$	Total Annual ³
Fixed-roof tanks	1.4	Int. floating roofs	1.0	17	0.6	4
Floating roof tanks ¹	0.2					
Loading emissions	0.2					
Point sources (C ² > 5%)	1.2	Class I	1.1	2.2	0.4	0.8
idem (C 0.5-5%)	0.7	Class II	0.6	1.1	0.2	0.2
idem (C 0.05-0.5%)	2.5	Class III	2.2	12	7.0	6.3
Point sources						
Flow rate < 50,000 m ³ /h C 0.05-0.5%	3.5	Class III				
C 50-200 (1000) m ³ /h	1.1	Class III	0.8	17	4.5	8
> 200,000 m ³ /h	1.4	Class III	1.1	17	4.5	8
Other sources	12.6	Increased maintenance and similar	6.4	28	20	25
Valves, flanges, etc. (2.6)						
Poorly defined (10.2)						

* This sector does not include manufacturing of rubber and plastic products.

Class I: Incineration, condensation and similar (low flow rates, high VOC concentrations), Class II: active carbon, biofiltration (high flow rates, low VOC concentrations). Class III, similar to class I, but decisions to take these measures pending.

¹) Total number of tanks with internal floating roofs about 400. From tanks mainly alkanes and aromatics are emitted. Alkenes and chlorinated hydrocarbons are for the larger part gaseous at ambient conditions.

²) C: concentration of VOCs in waste gases

³) Total annual costs is operating costs & 20% of investments. 1 US\$ = 1.8 Dfl.

Table 6.7 presents two types of measures as distinguished in the Dutch policy programme, those who will certainly be taken (the top three and the last measure) and so-called conditional measures, which will only be taken when specified conditions will be satisfied. These conditions are related to expected costs of measures and the expected VOC policy in other countries with competitive industries.

The data of Table 6.7 have been used to draw Figure 6.3, showing a marginal cost curve (emissions versus total annual costs of abatement (US\$ = 1,8 Dfl)). The marginal costs vary from US\$ 0.6 per tonne to US\$ 600 per tonne. Assuming the shape of Figure 6.3 is typical for European chemical industry, and for the time being, these data might be used to construct

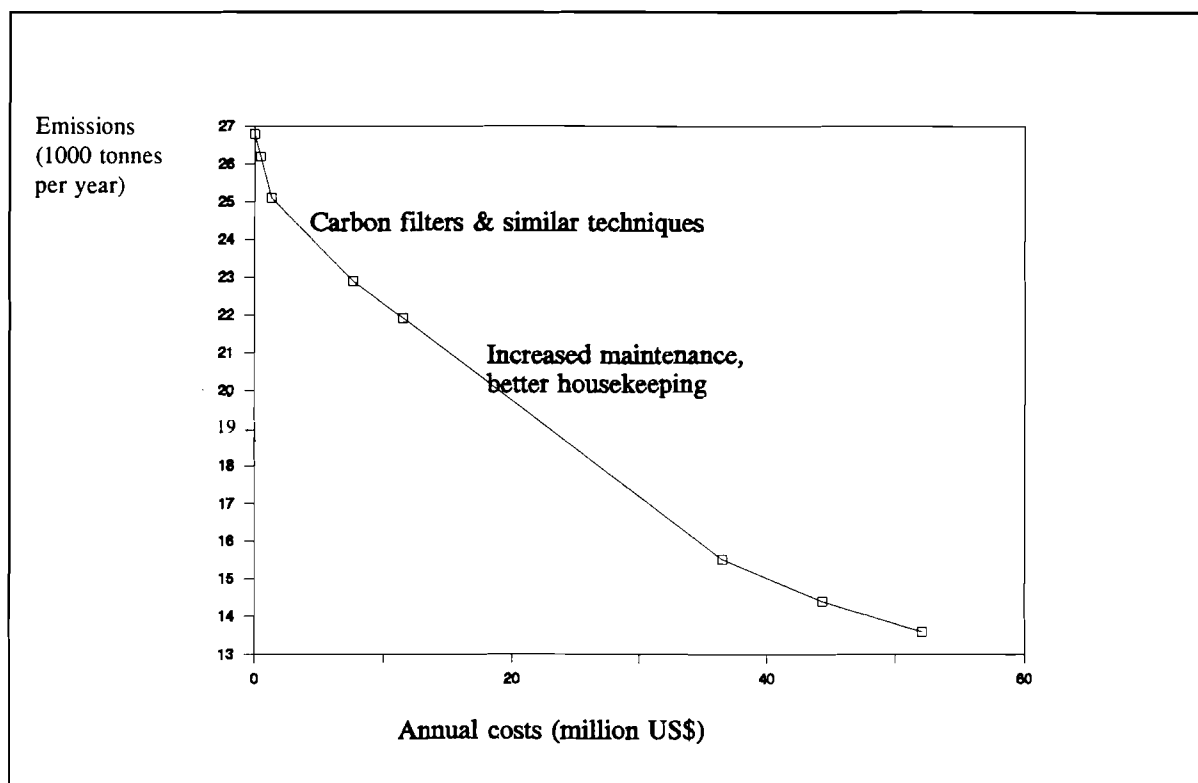


Figure 6.3 Cost curve for abatement of VOC emissions from the Dutch chemical industry (see Table 6.7).

similar cost curves for other countries provided that a key - such as emissions by country or production values - is available for up or down scaling. It is emphasized that this approach is very crude. Elaboration is possible by analysis of the chemical industries (emissions, structure, measures taken) in the various countries (or regions) which might lead to more appropriate cost curves (including some chemical speciation). However, this would require a considerable effort.

6.5 Solvent use

Industrial and building

From the UN-ECE inventory (Jourdan et al., 1990) the following industrial sectors appear as typical major VOC emitters:

- car industry
- appliances industry
- packaging and publishing printing
- textile industry

flatwood and wooden furniture industry
 architectural and other painting
 vegetable oil industry.

Except for the vegetable oil industry emissions are mainly due to surface coating operations.

Reduction of emission is possible by:

- good housekeeping practices (measures with negligible costs requiring mainly organisational efforts);
- improved equipment (such as electrostatic spray guns instead of pneumatic spray guns in car and appliances industry, higher freeboards in degreasing equipment);
- introduction of production processes which require less solvents or any solvent at all (such as low solvent and water-borne paints);
- application of add-on control techniques either to destroy or to recycle (in or out plant) organic compounds.

Experience shows that, in case of surface coating, a substantial reduction of VOC emissions should be possible by changing over to the use of water-borne paint (Olsthoorn et al., in prep.). Table 6.8 summarizes advantages and drawbacks of water-borne paints in surface coating in the production of metal furniture and other equipment as well as wooden furniture.

Table 6.8 Advantages and disadvantages of using water-borne paints for coating of metal and wooden surfaces (wooden and metal furniture and office equipment)

Advantages	Disadvantages
80-90% emission reduction	Costs of paint 10-15% up
Improvement of work conditions (health)	Increased use of chemicals in treating waste water
Less odour	Higher energy consumption
Less VOC imission	Extra investment in corrosion-persistent equipment
Paint easier to process	Extra pretreatment equipment
No fire risks	Might be less glossy
Less investment in storage facilities	
Less insurance premium	
No costs of thinner	

The current trend in coating technology is away from the use of organic solvents and

inefficient application techniques. Within the mix of incentives (economics, occupational health, fire-safety and environmental) behind this development, environmental policy increases in weight.

Table 6.9 Ranges of costs of abatement of VOC emissions due to the use of solvents

Sector/process	Cost range (US\$/tonne)
Metal degreasing	600 - 2500
Printing	300 - 2500
Car coating	2500 - 10000
Dry cleaning	500 - 2500
Coating of metallic, wooden, plastic surfaces	neg. - 2500
Impregnation of wood	1000 - 4500

Sources: Allemand, 1990, Giddings et al., 1991, Heslinga, 1990, Macdonald et al., 1991, Olsthoorn et al., in prep.

1 ECU= 1.3 US\$. Situation 1991/1992.

Economies-of-scale constitute an important factor in the explanation of the ranges given in Table 6.9. Development of cost curves, similar as in the case of the chemical industry, requires a compilation of data of country-specific frequency distribution of source strength (tonne/year) such as given in Table 6.10.

Table 6.10 Frequency distribution of emission sources (firms/plants) as found for the Dutch secondary metal industry (Thomas & Olsthoorn, 1987). Data relate to the period 1980-1982.

Emission class ton/year	Number of plants	Share in emission (%)
0 - 75	2000 - 3000	55
75 - 150	18	5
150 - 300	14	7
300 - 1000	7	8
1000 - 2000	4	15

From the data in the Tables 6.9. and 6.10 and data about emissions due to solvent use, it might be possible to estimate a country-specific cost curve, however, it appears wise to wait until data of better quality are available, considering the large uncertainties involved. Equally

important in deciding on elaboration are the requirements set by the atmospheric-chemical modelling.

Domestic solvent use

In households numerous products are used which contribute to VOC emissions. The most important products appear to be (see Table 5.3):

- paints and varnishes
- adhesives and glues
- cosmetics and personal care products
- polish and cleaners.

In this sector the control of emissions is obviously not possible by add-on measures. A decrease should be effected by promoting the use of non-VOC or low-VOC products, while putting a disincentive on the use of VOC-containing products or, perhaps, even a ban. Appropriate information about costs have not been found.

6.6 Combustion in stationary equipment

Emission control in the sense of end-of-pipe techniques is not considered as a viable option. The possibilities to decrease emissions are:

- reverting to other fuel. In particular abandoning the use of coal in domestic heating in favour of the use of natural gas. The emissions caused by combustion of coal in domestic stoves are orders of magnitude larger than emissions from natural gas combustion.
- energy conservation. Obviously, decreasing the demand for heat brings about less emissions.
- improvement of maintenance of domestic combustion equipment.

6.7 Other sectors

Control measures vary as widely as the sectors themselves.

6.8 Conclusions

The main area for control measures is road transport, in particular control of emissions from gasoline cars and control of emissions due to distribution of gasoline.

In the other areas less information is available, that is to say on a sectoral overall level¹⁵. Information on a detailed technical level is relatively abundant (Jourdan, 1990 and references). This information cannot be used in sector studies due to lack of statistical information about equipment and process applied. For instance a preliminary assessment of the costs of add-on control equipment requires data of flow rate (of off-gases containing the VOCs) and of the concentrations of the VOCs.

The assessment of the possibilities for VOC emission control outside the "gasoline sector" thus requires the elaboration of country-specific "technological profiles", sectoral characterizations enabling the use of the detailed technical information to construct cost curves representing possibilities for emission reductions and the costs involved.

7 CONCLUSIONS

This paper aims at presenting a concise overview of the availability of Europe-wide data about anthropogenic VOC emissions: sources, types of emitted VOCs, amounts emitted and how these might evolve over time, and VOC emission control measures (their cost and effectiveness). The background is IIASA's Transboundary Air Pollution Project, which aims at extending the RAINS model to study strategies for addressing transboundary tropospheric ozone pollution. The intended model should provide for scientific support for international negotiations within the framework of the UN-ECE Convention on Long Range Transboundary Pollution, similarly as the RAINS model does.

¹⁵ And with exception of the Netherlands in which case the policy plan to reduce emissions of VOCs is elaborated in sectoral scenarios with descriptions of goals, measures and their costs (KWS2000, 1989).

Only a few Europe-wide VOC emission inventories have been made, all used in the modelling of long-range tropospheric ozone formation. The CORINAIR methodology - originally developed for the EC - has evolved as the standard for Europe-wide inventorying; it was adopted by the UN-ECE task force on emission inventorying. Currently a Europe-wide inventory for the year 1990 is being made, using the CORINAIR90 system.

The results of this inventory will be used in the EMEP MSC-W¹⁶ long-range atmospheric pollution models, including its long period oxidant model. It is expected that the atmospheric modelling part of an integrated assessment model will be derived from the EMEP model. Loss of information in the derivation will be minimal when in the intended RAINS-type ozone model the same VOC emission inventory is used.

The main cause of man-made VOC pollution is traffic. Typically, about half of man-made VOC emissions are due to the use of gasoline. Solvent use ranks second as a source (about a quarter to a third). A characteristic of VOC emission estimates is their uncertainty. Emissions due to the use of solvents are particularly uncertain, in the range of -50% to +100%.

VOC emission estimating in inventorying is based for a large part on engineering judgment. This requires a knowledge of commonly used equipment and processes causing VOC emissions. This kind of information is available for North-Western European countries, however, decreases going south and in particular going east. Therefore, uncertainties in emission data, scenarios and possibilities for abatement are large in case of southern countries and in particular for the Eastern European countries.

Scenarios for the development of VOC emissions over time will differ sector by sector in credibility. Fortunately, having a major share in the emissions, the sector "road traffic" is among the sectors about which a credible forecast can be made, because such can be based on energy pathways - fuel consumption in transport - commonly used in other contexts.

¹⁶ EMEP: the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe, MSC-W ; Meteorological Synthesising Centre West, Oslo, Norway.

Unfortunately, solvent use is a "sector" about which information is inconclusive. The uncertainty in the magnitude of future emissions is large, not only because of uncertainty in current emissions but also due to trends (economic and technological) pointing in opposing directions. Therefore, lacking other information, keeping these emissions constant is a sufficiently good scenario.

This paper contains a survey of the possible measures to control VOC emissions. Aspects addressed are the areas of application, potentials of emission reduction and costs. Again information with respect to the traffic sector is relatively abundant. Currently equipment for the control of automotive emissions is introduced in Western Europe. In North-Western Europe measures are also taken, or expected, to control emission from the distribution of gasoline.

A large amount of information is available about techniques for both add-on VOC control and low-VOC emission processes, such as scrubbers, incineration and low-solvent or water-based paints. However, the level of this information is practically exclusively technical and detailed and not on a "sectoral" level. Development of sectoral information requires statistical analysis of processes and equipment used in sectors, comparable with sectoral information of type of boilers and their capacities which are used in energy-related studies. At present, such information is scarce, however, with the development of the UN-ECE emission inventory country-specific statistical information will become available and will facilitate the construction of a RAINS-type of ozone model.

VOC is a collective for a large group of chemical compounds. The contributions of the compounds to ozone pollution diverge: some compounds have only local influence, others contribute to trans-boundary pollution, while an organic compound like methane has no spatially distributed effects. These differences are due to diverging chemical reaction rates which differ with type of VOC. The photochemical models used today take account of these differences in a simplified way.

Sources usually emit mixtures of VOCs; it occurs only rarely that a single VOC is emitted. Therefore, a control measure usually affects the emissions of different VOCs simultaneously.

The concept of cost curves, similar as used in RAINS, cannot exactly be adopted in the ozone case. In the former case these curves are two-dimensional (costs versus emissions), whereas in the ozone case the curve is expanded to a surface with n-dimensions (emissions, costs, type of VOC). To what extent such refinement will be made eventually depends on uncertainties in information about both emissions and control measures and requirements set by the atmospheric model within the conceived model.

This paper basically addresses the VOC emission module: available data and the data format. The work on the atmospheric modelling is yet to begin. The results of this work, together with the expected improved emission inventories, will give the guidance for further elaboration of the VOC emission module.

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