

EU ENVIRONMENT DIRECTORATE

**PHOSPHATES AND ALTERNATIVE DETERGENT
BUILDERS – FINAL REPORT**

**WRc Ref: UC 4011
June 2002**

PHOSPHATES AND ALTERNATIVE DETERGENT BUILDERS – FINAL REPORT

Report No.: UC 4011

31 May 2002

Authors: E B Glennie, C Littlejohn, A Gendebien, A Hayes, R Palfrey, D Sivil and K Wright

Contract Manager: A S Dee

Contract No.: 12565-0

RESTRICTION: This report has the following limited distribution:

External: EU Environment Directorate

Internal: Authors

Any enquiries relating to this report should be referred to the authors at the following address:

WRc Swindon, Frankland Road, Blagrove, Swindon, Wiltshire, SN5 8YF.
Telephone: + 44 (0)1793 865000 Fax: + 44 (0) 1793 865001

The contents of this document are subject to copyright and all rights are reserved. No part of this document may be reproduced, stored in a retrieval system or transmitted, in any form or by any means electronic, mechanical, photocopying, recording or otherwise, without the prior written consent of the copyright owner.

This document has been produced by WRc plc.

CONTENTS

SUMMARY	1
1. INTRODUCTION	6
1.1 Background	6
1.2 Role of phosphorus in surface waters	7
1.3 European perspective	10
1.4 Project aim	11
1.5 Project Objectives	11
2. DETERGENT BUILDERS AND DETERGENT USE	12
2.1 Constituents of detergents	12
2.2 Types of detergent	17
2.3 Current detergent use in Europe	17
3. CASE STUDIES OF ACTIONS TAKEN TO LIMIT OR BAN PHOSPHATES IN DETERGENTS	21
3.1 Review of action to date	21
3.2 Walloon Region of Belgium	31
3.3 France	40
3.4 Germany	49
3.5 Hungary	54
3.6 Italy	58
3.7 Netherlands	65
3.8 Conclusions	69
3.9 Switzerland	70
3.10 The USA	74
4. DETERGENT ECOLABEL SCHEMES	83
5. THE PHOSPHATE & ZEOLITE INDUSTRIES IN EUROPE	85
5.1 STPP Production	85
5.2 Phosphate rock extraction and phosphate manufacturing processes	86
5.3 Phosphoric acid manufacturing processes	88
5.4 Manufacture of Sodium Tripolyphosphate	90
5.5 European STPP manufacturers	91
5.6 Zeolite A manufacturers in Europe	93
5.7 Conclusions	93
6. DISCHARGES OF PHOSPHORUS TO SURFACE WATERS	94
6.1 Industrial discharges of phosphorus	94
6.2 Agricultural inputs of phosphorus	94
6.3 Municipal wastewater	95
7. LIFE CYCLE ANALYSIS	102

7.1	Introduction	102
7.2	Processes for phosphorus removal from wastewater	102
7.3	Detergent builders – STPP	111
7.4	Detergent builders – Zeolite A	117
7.5	Detergent builders – Polycarboxylates	119
7.6	Comparison between detergent builders	119
8.	CONCLUSIONS AND RECOMMENDATIONS	121
8.1	Overall Conclusions	121
8.2	Recommendations:	122
8.3	policy options for controlling phosphorus	125

APPENDICES

APPENDIX A	REFERENCES	127
APPENDIX B	AGRICULTURAL AND INDUSTRIAL SOURCES OF PHOSPHORUS	131
APPENDIX C	PHOSPHORUS DISCHARGES TO SURFACE WATER FROM MUNICIPAL WASTEWATER	145
APPENDIX D	USA NATIONAL WATER QUALITY INVENTORY	151
APPENDIX E	COST AND ENERGY MODEL OF WASTEWATER AND SLUDGE TREATMENT	157
APPENDIX F	SLUDGE PRODUCTION ESTIMATES	165
APPENDIX G	ZEOLITE A	171

LIST OF TABLES

Table 1-1	JRC classification of trophic level	9
Table 2-1	Substances used in detergents	13
Table 2-2	Comparison of typical P based and P free Laundry Detergent Formulations (Conventional Powders)	14
Table 2-3	Typical Laundry Detergent Formulations (<i>Compact Powders</i>)	15
Table 2-4	Constituents of some detergents	15
Table 2-5	Estimated detergent consumption in Europe with current legislation	19
Table 3-1	Legislative and Voluntary Frameworks for Phosphates in Detergents	22
Table 3-2	Trends in STPP consumption	28
Table 3-3	Type of WWT plants in the Walloon Region	32
Table 3-4	List of WWTP in the Walloon Region with P removal	33
Table 3-5	Treatment efficiency and nutrient loading from WWT plant (tonne per year)	38
Table 3-6	Nutrient load from existing and future sewerage network* (tonne per year)	38
Table 3-7	Nutrient load from individual habitat (tonne per year)	39

Table 3-8	Nutrient load from direct discharge from industries	39
Table 3-9	Diffuse nutrient losses (tonnes per year)	39
Table 3-10	Summary of P inputs to river systems	40
Table 3-11	Estimated proportions of total P removed in sewage treatment: France	43
Table 3-12	Estimates quantities of P discharged to German rivers (Hamm)	50
Table 3-13	Estimates quantities of P discharged to German rivers (Behrend et al) 51	
Table 3-14	Wastewater collection and treatment levels – Hungary, 2001	56
Table 3-15	Sources of Phosphorus in the Danube Basin	56
Table 3-16	Estimated quantities of total P from population discharged to Hungarian surface waters, 2010	57
Table 3-17	Lake Endine history	62
Table 3-18	Phosphate (total-P) pollution of surface water in the Netherlands, 1985 – 1995, in 1000 ton/year (source: RIZA)	67
Table 3-19	Summary of Development of Legislation in Switzerland	71
Table 3-20	Summary of USA policy development and legislation	75
Table 3-21	USA state bans on STPP in detergents	78
Table 5-1	World production of phosphate, 1995 - 1999	86
Table 5-2	World uses of phosphate	86
Table 5-3	European STPP manufacturers	91
Table 5-4	Examples of products that contain phosphorus	92
Table 5-5	Estimates of detergent builder use in Europe	92
Table 6-1	Phosphorus flows – agriculture Switzerland 1994	94
Table 6-2	Per capita detergent use	96
Table 6-3	Estimates of phosphorus discharged to sensitive areas	100
Table 6-4	Population in small centres for some major catchments	101
Table 7-1	Wastewater and sludge treatment processes used for the LCA comparison	103
Table 7-2	Comparison of the treatment processes	106
Table 7-3	Pros and cons of chemical P removal	107
Table 7-4	Pros and cons of biological P removal	108
Table 7-5	Model outputs for process option 2A, sludge to agricultural land	109
Table 7-6	Sludge production in biological sewage treatment	110
Table 7-7	Impacts of STPP production	113
Table 7-8	STPP production – ThermPhos process	115
Table 7-9	STPP production – wet process	116
Table 7-10	Impacts of Zeolite A production	117
Table 7-11	Zeolite A production processes	118
Table 7-12	Impacts of polycarboxylate production	119
Table 7-13	Comparison between STPP and Zeolite A	119
Table 8-1	Summary of river catchment case studies	123
Table 8-2	Summary of lake case studies	124

Table B.1	Land use by country	132
Table B.2	Phosphorus fertiliser consumption per unit area of agricultural land by country (FAO 2001)	133
Table B.3	Phosphorus inputs – specific cases	143
Table C.1	Municipal Wastewater Treatment – Current Situation	145
Table C.2	Population by size of centre	146
Table C.3	Assumed P discharged for different treatment types	146
Table C.4	Future, UWWTD compliant, wastewater treatment	147
Table C.5	Phosphorus discharges under different scenarios	148
Table C.6	Phosphorus discharges to sensitive areas – selected countries	149
Table E.1	Wastewater and sludge treatment processes modelled	157
Table E.2	Process model assumptions	158
Table E.3	Process model results, 12 mg/l P in crude sewage	159
Table E.4	Process model results, 8 mg/l P in crude sewage	160
Table E.5	Process model results, 15 mg/l P in crude sewage	161
Table E.6	Process model results, 12 mg/l P in crude sewage, P availability in sludge 50%	162
Table E.7	Process model results, 12 mg/l P in crude sewage, sidestream P availability 50%	163

LIST OF FIGURES

Figure 1.1	Biochemical Phosphorus Cycle	8
Figure 3.1	Trends in domestic P-free laundry detergent in Belgium (DETIC, pers com 2001)	32
Figure 3.2	Comparison of median concentrations for Tot P, Meuse	35
Figure 3.3	Concentration in Chlorophyl a, Meuse	36
Figure 3.4	Total P concentrations, Schelde	37
Figure 3.5	Chlorophyll <u>a</u> concentration, Schelde	37
Figure 3.6	Total P concentrations in 4 French rivers	45
Figure 3.7	Concentrations of orthophosphate and total phosphate in Rhine water at Lobith, 1975-1998 (source: RIWA, 2000)	52
Figure 3.8	Total P trend in the IJsselmeer (Source: ETC/IW)	68
Figure 3.9	Total phosphorus concentrations monitored in the River Meuse at Keizersveer, 1977-1995 (Source: Data as reported to ETC-Inland Waters)	68
Figure 3.10	Total phosphorus concentration in Lake Geneva, 1957-1995	72
Figure 3.11	Phosphate limits in US States (1971-1995)	77
Figure 5.1	Crude acid purification	89
Figure 5.2	STPP production	90
Figure 6.1	Discharges of phosphorus to surface water: France	98
Figure 6.2	Discharges of phosphorus to surface water: Portugal	98

Figure 6.3	Discharges of phosphorus to surface water: Spain	99
Figure 6.4	Discharges of phosphorus to surface water: UK	99
Figure 6.5	Discharges of phosphorus to surface water: Poland	100
Figure 7.1	Chemical Phosphorous Removal for 20,000pe works	104
Figure 7.2	Biological and Chemical Phosphorous Removal for 200,000pe works	105
Figure B.1	Phosphorus fertiliser consumption in Europe	134
Figure B.2	Cattle numbers, 1990-2000 – EU and accession states	136
Figure B.3	Chicken numbers (000s) 1990-2000 – EU and accession states	137
Figure B.4	Pig numbers 1990-2000 – EU and accession states	138
Figure B.5	Sheep numbers 1990-2000 – EU and accession states	139

Glossary of acronyms

AI SD	Association des Industries de Savons et des Détergents (International Soap and Detergent Association)
AISE	Association Internationale de la Savonnerie, de la Détergence et des Produits d'Entretien (International Association for Soaps, Detergents and Maintenance Products)
AS	Activated sludge
BOD	Biochemical Oxygen Demand
BPR	Biological Phosphorus Removal
CEC	Commission of European Communities
CED	Comité Environnement Détergents
CEE	Central East-European
CEFIC	Conseil Europeen des Federations de l'Industrie Chimique (EDI Project for Chemical Industry)
CESIO	European Committee on Organic Surfactants and their Intermediates
CIPM	Comité International des Poids et Mésures
CMC	Carboxymethylcellulose
CMOS	Carboxymethyloxysuccinate
CMT	Carboxymethyltratronate
CNR	Consiglio Nazionale delle Ricerche
COD	Chemical Oxygen Demand
CWA	Clean Water Act
DETIC	Belgian-Luxembourg Association of Manufacturers and Traders of soaps, detergents, maintenance products, cosmetics, adhesives and similar products
DETR	UK Department of the Environment, Transport and the Regions
DG	Director General
DGRNE	General Division for Natural Resources and Environment
DRBC	Delaware River Basin Commission
EAWAG	Swiss Federal Institute for Environmental Science and Technology
EBRD	European Bank for Reconstruction & Development
EC	European Commission
EDF	European Development Fund
EDTA	Ethylenediaminetetracetic acid
EEA	European Environment Agency
EEC	European Economic Community
ELVs	Emission limit values
EMPA	Eidgenössische Materialprüfungs und Forschungsanstalt (the Swiss Federal Laboratories for Materials Testing and Research)
EPDRB	Environmental Programme for the Danube River Basin
ETC	European Topic Centre on Water

ETC-IW	European Topic Centre - Inland Waters
EU	European Union
EUEB	European Union Eco-labelling Board
FAO	Food and Agriculture Organization (United Nations)
FAOSTAT	FAO Statistical Database
FEM	French Environment Ministry
FMF	French Ministry of Finance
FSU	Former Soviet Union
GJ	Gigajoule
HELCOM	Convention on the Protection of the Marine Environment of the Baltic Sea Area (The Helsinki Convention)
ICPR	International Commission for the Protection of the Rhine
IDAPA	Irish Detergent Industry Association
IFEN	Institut francais de l'environnement
IKSR	Internationale Kommission zum Schutze des Rheins
ISBN	International Standard Book Number
ISTAT	Istituto Centrale di Statistica (Italian National Statistics Institute)
LAS	Linear alkyl benzene sulphonate
LCA	Life Cycle Assessment
LOICZ	Land-Ocean Interactions in the Coastal Zone
MAFF	Ministry of Agriculture, Fisheries and Food (UK)
N	Nitrogen
NERI	National Environmental Research Institute, Denmark
NPDES	National Pollutant Discharge Elimination System
NPE	Net primary energy
NTA	Nitrilotriacetic acid
NVZ	Dutch Soap Association
OECD	Organisation for Economic Cooperation & Development
OSPAR	The Convention for the Protection of the Marine Environment of the North-East Atlantic
P	Phosphorus
PCAs	Polycarboxylic acids / Polycarboxylates
PCS	Permit Compliance System
PEC	Predicted Environmental Concentrations
PNEC	Probable No Effect Concentrations
POTW	Publicly Owned Treatment Works
RAP	Rhine Action Plan Against Chemical Pollution
RAS	Return Activated Sludge
RIVM	National Institute of Public Health and Environmental Protection (Netherlands)
RIWA	Vereniging van RivierWaterbedrijven, Netherlands (Association of River

	Waterworks)
RIZA	National Institute of Inland Water Management and Waste Water Treatment (Netherlands)
RNDE	French Water Data Network
SAS	Surplus Activated Sludge
SCOPE	Scientific Committee On Problems of the Environment
STPP	Sodium tripolyphosphate
TGAP	Taxe Générale sur les Activitiés Polluantes (General Tax on Polluting Activities)
TSS	Total suspended solids
UBA	German Umweltbundesamt
UK	United Kingdom
USA	United States of America
USEPA	USA Environment Protection Agency
USGS	United States Geological Survey
UWWT	Urban Wastewater Treatment
UWWTD	Urban Wastewater Treatment Directive
VFAs	Volatile fatty acids
VRM	Dutch Ministry of Housing, Spatial Planning and the Environment
WPCF	Water Pollution Control Federation
WRc	Water Research Centre (WRc plc)
WWTP	Wastewater Treatment Plant
ZEODET	Association of Detergent Zeolite Producers

SUMMARY

Introduction

Recognition of the relationship between increasing phosphorus inputs to surface waters and the subsequent increase in eutrophication of water bodies gave rise to public concern during the 1970's and 1980's. This led to action by several countries including the USA, Japan and some EU member states, to reduce phosphorus loads, particularly from urban and industrial point sources.

The two main areas of action that have taken place, particularly in the late 1980's and early 1990's are:

- A reduction in the amount of sodium tripolyphosphate (STPP) used in detergent builders and switch to 'alternative' non-phosphate based builders, such as Zeolite A; and,
- Improving wastewater treatment through implementation of the Urban Wastewater Treatment Directive (UWWTD).

Where STPP is used as builder in household detergents it contributes to up to 50% of soluble (bioavailable) phosphorus in municipal wastewater, therefore a reduction in the use of phosphate based detergents should have a positive impact on the eutrophication of surface water bodies. Measures to reduce the use of STPP based detergents in the EU included the introduction of laws or voluntary agreements to change to Zeolite A as the builder for household laundry detergents. As a result STPP consumption has decreased substantially since the early 1980's, with dramatic decreases observed in Germany, Italy, the Netherlands and Switzerland. The widespread introduction of zeolite based detergents, even in countries where no formal action was taken, implies widespread acceptance of zeolite based detergents throughout Member States.

The European Commission (EC) has implemented this study to address the current use of phosphates in detergents throughout the European Union (EU) and recommend appropriate measures to improve the current situation. The study covers the fifteen Member States of the EU and the three accession countries Poland, Hungary and the Czech Republic.

The aim of the study is to investigate the costs and benefits of substituting phosphorus in detergents with other appropriate builders and to provide recommendations on the most appropriate method of reducing phosphorus concentrations in surface waters, through either improving wastewater treatment, banning the use of phosphates as detergent builders, or a combination of the two approaches.

Measures to reduce or ban phosphates in detergents

Detailed case studies were undertaken for eight countries, five of which are EU Member States and one Accession State. These are:

- Belgium (Walloon Region);
- France;
- Germany;

- Hungary;
- Italy;
- The Netherlands;
- Switzerland; and,
- The USA.

The case studies provide an overview of the voluntary and legislative measures that have been introduced in each country to limit the use of phosphorus based detergents and improve wastewater treatment facilities. The case studies then provide an assessment of the impacts that these measures have had on reducing phosphorus concentrations and subsequent eutrophication of surface waters.

With the exceptions of Belgium and the Irish republic, measures to move from the use of STPP to Zeolite A in domestic laundry detergents in EU member states were initiated by 1990. Most measures were either statutory limits on the STPP content, or voluntary agreements with detergent suppliers.

As a result of these measures STPP consumption decreased dramatically between 1984 and 1990 in Germany, Italy, the Netherlands and Switzerland, and is now effectively zero in these countries. In all these countries, voluntary or legislative action was taken during the same period. STPP consumption decreased more gradually between 1984 and 1990 in Austria, Belgium, Denmark, Finland, Ireland and Sweden, although is now low or zero. In other EU member states, household laundry detergents built from STPP and from Zeolite A have roughly equal market shares, including France, Greece, Portugal, Spain, UK. The same applies in the Czech Republic and Hungary. However, in Poland, most household laundry detergents sold are built from STPP.

The phosphate and zeolite industries in Europe

An overview of the phosphate and zeolite industries in Europe is made, including details of production, extraction and manufacturing processes.

The two distinct components to the phosphate industry in Europe are the fertiliser and chemical industries. While the fertiliser industry requires lower levels of phosphate purity, the quantity of phosphorus used is 10 times that of STPP. The chemicals industry supplies foods, detergents and a variety of other industries, of which over 50% of non-fertiliser phosphate is used for detergents.

The European STPP production industry is relatively small, contributing to less than 10% of overall world production. China and India are major producers. A ban on STPP use in detergents in the EU would be likely to reduce the European STPP manufacturing base, and increase the risk of production being moved elsewhere in the world.

In comparison, approximately 50% of detergent zeolites are produced in Europe, the capacity for production exceeds current production, and it is likely that any increased demand for Zeolite A could be met without any additional major investment.

Discharges of phosphorus to surface waters

Estimated quantities of phosphorus discharged to surface water via municipal households are presented and the current situation compared to a number of scenarios, namely:

- i. If there were a complete ban of STPP use;
- ii. Full implementation of the UWWTD; or,
- iii. A combination of i & ii

While industrial sources may be important locally, the two main sources of phosphorus inflows to surface water are municipal wastewater and agriculture. In catchments with low levels of wastewater treatment (i.e. no P removal) municipal wastewater generally represents the largest source of phosphorus. However, where municipal wastewater treatment is of a high standard (e.g. tertiary with P removal), the largest source of phosphorus is from agricultural inputs.

The main agricultural sources are from animal husbandry or fertiliser use, with erosion and run off being the major transport pathways of phosphorus to surface waters.

Phosphorus from detergents contributes an estimated 25% of phosphorus in municipal wastewater requiring treatment in the EU Member States where STPP is still used, Hungary and the Czech Republic. However, the percentage is likely to be higher in Poland, where most detergents are built on STPP.

Phosphorus discharges are reduced considerably by both banning STPP from detergents and improvements to wastewater treatment. However, their combined effect is less than the sum of the individual effects. Even following full implementation of the UWWTD, significant quantities of phosphorus would still be discharged to surface waters, from dispersed populations and population centres less than 10,000, and in non-sensitive areas.

Life Cycle analysis

A life cycle comparison between STPP and Zeolite A based detergent builders is provided, for two wastewater treatment options; one using chemical phosphorus removal and the other using biological phosphorus removal.

No distinction is made between STPP and Zeolite A in terms of the cost of detergents to householders or their cleaning efficiency. There is some evidence from consumer magazine surveys that STPP is preferred. However zeolite based detergents are sold successfully in supermarkets alongside STPP based detergents in countries such as the UK and France where both are freely available.

No major differences were observed in the production energy requirements per kg builder, environmental impacts and sludge production between STPP and Zeolite A, and neither were shown to be toxic to aquatic fauna.

Overall Conclusions and Recommendations

A number of countries have been successful in reducing eutrophication through implementation of measures to reduce phosphorus loads. Notable examples are Lake Geneva in Switzerland, Lake Erie in the USA and Lake Endine in Italy. In all cases the results

indicate that a phosphorus reduction of 70%-90%¹ is necessary to significantly reduce eutrophication and improve trophic status.

A ban on the use of phosphate based detergents can achieve a phosphorus load reduction of up to 40% entering surface water bodies, which is not sufficient in isolation to result in any substantial improvements. Furthermore, improvements in wastewater treatment to fully comply with the UWWTD would only result in typical phosphorus reductions of around 30%. As demonstrated by Switzerland, the USA and Italy, the greatest improvements in lakes and rivers were observed where a combination of reduced detergent phosphorus and improved wastewater treatment were implemented, thereby achieving the required 70-90% reduction in external load.

The main sources of phosphorus entering surface waters are from municipal wastewater and agriculture. However, relative contributions vary depending on the nature of catchment landuse activities. For example, in areas without intensive agriculture (lake Geneva's catchment, lake Endine), municipal wastewater is the major source of phosphorus and in these areas improved wastewater treatment has been effective in reducing eutrophication. On the other hand, in catchments with intensive agriculture (e.g. lake Sempach in Switzerland, Wallonia, lower Rhine), agricultural inputs of phosphorus may represent a major source and a combination of measures including improved wastewater treatment and adoption of best land management practices should be employed.

Although the full implementation of the UWWTD will result in substantial reductions in phosphorus loads, discharges of wastewater without phosphorus removal would continue in sensitive areas, where the population is dispersed or in centres up to 10000 population equivalents. Further action to reduce phosphorus loads entering surface waters may be required in these areas.

Based on the results of life cycle analysis, Zeolite A was found to be a suitable alternative to STPP for use as a detergent builder. Only minor differences were observed in overall production cost in terms of energy used and sludge produced. Additionally, Zeolite A was found to be non toxic to aquatic fauna and humans and produces less toxic waste by-products when extracted from bauxite than phosphorus containing rocks (e.g. tailings produced include the heavy metals quantities are relatively minor). Furthermore, Zeolite A based detergents is generally accepted by EU Member States and consumers as an efficient and acceptable alternative to STPP based ones. The life cycle analysis concluded that 'any decision on the selection of a detergent builder should be based on other factors'.

The EU contributes to less than 10% of the world's STPP production, and employs approximately 1000 people. Therefore, while an EU wide ban on STPP use would direct STPP manufacturing to other large centres, such as China and India, the economic loss of this is not considered to be great in overall EU terms. Additionally, as the current EU capacity for Zeolite A production exceeds the actual production, it could be expected that increased production in this area would result in substantial employment and economic opportunities, with the only a small requirement for additional capital expenditure on infrastructure.

Excessive amounts of phosphorus has long been implicated in the eutrophication of surface water bodies. Therefore, to promote lake/river recovery and improve trophic status it is imperative that phosphorus loads entering surface waters are reduced. Based on the analysis

¹ Compared to 100% STPP based detergents and no nutrient removal from wastewater

of a number of countries, this phosphorus load reduction should be greater than 70% in order to achieve the above objectives. This can only be achieved through the implementation of a combination of limiting/banning the use of STPP based detergents and improving waste water treatment.

Zeolite A was shown to be a cost-effective alternative, both in terms of socio-economic and environmental impacts, to the use of STPP as a detergent builder in the EU. Therefore measures should be employed on an EU scale to restrict/ban the use of STPPs and switch to detergent builders based on Zeolite A.

Recommendations:

Based on the conclusions outlined above, the following recommendations are made:

- That a general ban on the use of STPP as a builder for household detergents be placed on all EU Member States;
- That EU Member States endeavour to reduce phosphorus loads entering surface waters in order to reverse the long term trend of eutrophication, through a combined approach of banning STPPs in household detergents and achieving full implementation of the UWWTD;
- That further investigations are undertaken on scattered populations and centres less than 10000 equivalents to determine the relative phosphorus contributions originating from these sources, after full implementation of the UWWTD, and what measures are needed and could be employed to reduce these contributions;
- That further investigations be undertaken within agricultural areas to identify 'best management practices', to reduce phosphorus loss to surface waters.

1. INTRODUCTION

1.1 Background

High and rising levels of phosphorus in surface waters in the 1970s, and the increased occurrence of eutrophication, gave rise to public concern on the possible causes. One of the main sources was identified to be the use of phosphorus in household detergents. In several countries, including the USA, Germany, Italy and Switzerland this concern led to action to reduce the amount of phosphorus entering surface water bodies, through either improved waste water treatment or the removal of phosphorus based detergents.

There was widespread debate on the merits of substituting laundry detergents built from sodium tripolyphosphate (STPP) with those built from Zeolite A or other alternatives. The parties in the debate included voluntary environmental groups, governments and commercial interests: suppliers of STPP and of Zeolite A, and industries such as tourism and fisheries that were adversely affected by eutrophication.

With the exceptions of recent measures in Belgium and the Irish Republic, the measures in all Member States of the European Union (EU) were initiated by 1990.

In some countries the debate resulted in laws or voluntary agreements to change to Zeolite A as the builder for household laundry detergents. In others there has been a partial change, and the debate continues.

Most measures on detergents were either statutory limits on the STPP content, or voluntary agreements with detergent suppliers to supply only zeolite based detergents. Legal bans have been applied in 5 countries considered here, one of them in the EU.

- Canada (1973)
- Italy (1989)
- Japan. (Ban limited to areas containing sensitive lakes but in effect no STPP based detergents are used in Japan).
- Switzerland (1986).
- USA (different dates in different states from the 1970s onwards).

STPP consumption decreased dramatically between 1984 and 1990 in Germany, Italy, the Netherlands and Switzerland. In all these countries, voluntary or legislative action was taken during the same period.

In most other countries there was a steady downward trend in STPP consumption, and corresponding penetration of the market by zeolite based detergents. This penetration has occurred throughout the EU, including countries where no formal action was taken, such as France, Greece and the UK. This implies widespread acceptance of zeolite based detergents.

The other major impact on the reduction of phosphorus in surface waters has been the recognition of the need for improved sewage treatment, and the subsequent implementation of the Urban Wastewater Treatment Directive (UWWTD) which entered into force in 1991.

This study has been implemented by the European Commission to address the current use of phosphates in detergents throughout the European Union (EU) and recommend appropriate measures to improve the current situation. The study covers the fifteen Member States of the EU and the three accession countries Poland, Hungary and the Czech Republic.

This report represents the final outcomes of the study.

1.2 Role of phosphorus in surface waters

Phosphorus enters surface water bodies via non-point sources such as agricultural runoff and animal husbandry, and from point source municipal and industrial wastewater discharges. The relative importance of these sources varies widely between catchments, depending on:

- the degree of urbanisation;
- the standard of sewage treatment; and,
- the nature and intensity of agricultural practices (i.e. whether animal husbandry or vegetable crops).

Industrial sources are considered to contribute a smaller overall load to surface waters than either agriculture or municipal wastewater.

In catchments where household laundry and dishwasher detergents contain phosphate as a builder, up to 50% of soluble phosphorus in municipal wastewater comes from this source.

Nutrients, particularly nitrogen and phosphorus, are essential elements used in plant and algal metabolism and therefore integral in influencing the productivity of freshwaters. While many other elements contribute to the metabolic synthesis of fats and proteins, phosphorus is generally considered to be the primary nutrient limiting aquatic plant growth, and is the key nutrient implicated in the eutrophication of fresh waters (Vollenweider 1976, Twinch 1986). The majority of phosphorus in freshwaters occurs as organic phosphates, with about 70% retained in living or dead biomass and the remainder as either soluble or particulate phosphorus. Soluble phosphorus (orthophosphate) is the main bioavailable form of phosphorus (Wetzel 1983).

The majority of phosphorus enters natural waters in a non-bioavailable form, bound to particulate matter, with only around 5% occurring in soluble form. However, soluble phosphate in sewage effluent can be as high as 90% and may alter the balance of particulate and dissolved phosphate input to surface waters, particularly in highly impacted catchments (Wetzel 1983). The key elements of the biochemical phosphorus cycle are shown in Figure 1.1.

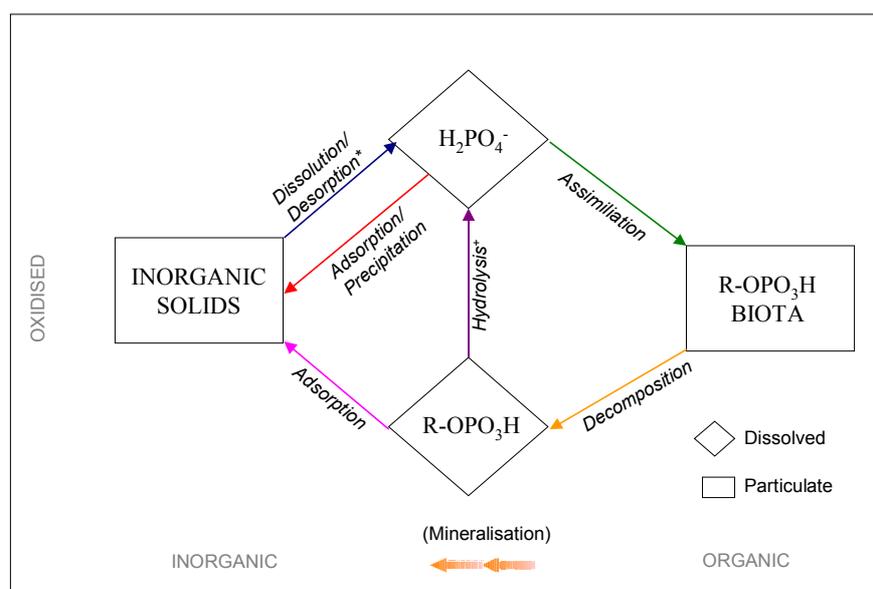


Figure 1.1 Biochemical Phosphorus Cycle

Note: Figure 1.1 shows the major reactions occurring between organic and inorganic states (+ = enzymatic, photochemical, pH variability; * = reductive, photochemical, pH variability)

1.2.1 Phosphorus cycling in surface waters

The importance of sediments in the cycling of phosphorus is widely acknowledged. While there is generally a net flux of phosphorus to the sediments each year, re-mobilisation of soluble phosphorus from the sediment can occur under certain conditions. Phosphorus exchange across the sediment-water interface is influenced by oxygen concentrations and redox reactions, pH, ion complexation and activities of benthic flora and fauna (Gachter and Meyer 1990, Gonsiorczyk *et al.* 1997).

Phosphorus concentrations in sediments are generally much greater than those of the overlying water. Soluble phosphate is released from sediments into the overlying water when dissolved oxygen concentrations fall below 2 mg/L (Gachter and Wehrli 1998, Mortimer 1941 & 42). The rate of release can be up to 1000 times faster in anoxic waters than under oxygenated conditions (Horne and Goldman 1994). However, this rate of release is dependant on such factors as the adsorption/desorption capacity of the sediment, the conditions of the overlying water, and the composition of organic carbon and biota within the sediment (Gachter and Meyer 1990).

Phosphorous forms complex bonds with numerous metal oxides, such as ferric iron, manganous manganese, zinc and copper. The binding capacity of phosphorus to these metal oxides is strongly dependent on the redox conditions at the sediment-water interface. In oxygenated waters, phosphorus is readily bound to iron oxides. Alternatively, under anaerobic conditions ferric iron is reduced leading to the release of soluble phosphate.

In waterbodies where phosphorus concentrations and water residence times are sufficient to cause oxygen depletion, significant amounts of soluble phosphate can be remobilised from the sediments. Therefore, due to the low water residence times and elevated oxygen levels, remobilisation of soluble nutrients from the sediments is generally considered to be low in flowing waters (e.g. rivers and estuaries). However, in large or slow flowing rivers (e.g. lowland), residence times may be sufficient to deplete oxygen resources, thereby facilitating the release of dissolved nutrients from the sediments.

1.2.2 Lake trophic status and phosphorus buffering capacity

Internal phosphorus cycling is influenced by trophic status, as different trophic levels create different conditions for lake metabolism. Oligotrophic lakes are resilient to increases in nutrient loading and can retain large amounts of phosphorus in the lake sediments. This is mainly due to the high **phosphorus buffering capacity** of the sediments, which is the equilibrium between soluble and particulate phosphorus (Twinn 1986). At the onset of eutrophication, phosphorus concentrations in the water column remain low in relation to the external load, as phosphorus bound to particulate matter is sedimented. With prolonged phosphorus inputs, the buffering capacity of the sediment is exceeded resulting in large phosphorus concentrations in the water column.

Phosphorus residence time in lakes is strongly related to trophic condition. Furthermore, the resilience of lakes will depend on their previous history, in that oligotrophic lakes will respond slowly to an increased load and quickly to a decreased load, while eutrophic lakes will respond quickly to an increased load and slowly to a decreased load. An example of lake recovery following a short period of enrichment has been demonstrated by Holmgren (1984) who fertilised four lakes in northern Sweden over a period of four years. While the nitrogen and phosphorus enrichment resulted in a 50-60% increase in algal biomass, this returned to normal within one year of ceasing the experiment. Alternatively, delays in recovery of eutrophic lakes with a longer history of enrichment, following a reduction in external load has been shown in a number of lakes (e.g. Upper Kis-Balaton Reservoir - Hungary, Lake Sempach – Switzerland, Lake Trummen – Sweden, Lake Shagawa – USA, Lake Asvalltsjarn – Sweden, Lake Sheelin - Eire). This prolonged delay can extend for many years. For example Lakes Asvalltsjarn and Sheelin showed no change in trophic status over a period of ten years (Marsden, 1989).

The European Commission, Joint Research Centre (JRC) have designated five trophic classes of multiple use lakes, using concentrations of total phosphorus (Table 1.1). These classes have been adapted from the OECD boundary values for trophic classification following assessment of the existing criteria used in some Member States for freshwater subject to eutrophication (Cardoso *et al*, 2001).

Table 1-1 JRC classification of trophic level

Class	Trophic Level	Total P ($\mu\text{g/L}$)
1	Oligotrophy	<10
2	Oligo-mesotrophy	<20
3	Mesotrophy	<50

4	Eutrophy	<100
5	Hypertrophy	>100

1.3 European perspective

The trophic status of a water body will tend towards equilibrium with its catchment, so that reductions in external phosphorus loading will eventually result in a reduction in receiving water phosphorus concentrations. However, the extent to which a reduction in load reduces surface water phosphorus concentrations is influenced by morphometry, flushing rates, sediment types, trophic status and the history of enrichment. Historical land-use pressures within a river basin will strongly influence trophic status. Therefore, it would be expected that a water body in a highly modified catchment, with significant agricultural or industrial development, or low levels of wastewater treatment, would be more enriched than a water body with few catchment impacts. Such impacted water bodies are expected to respond more slowly to a reduction in external phosphorus loads, due to the large pools of phosphorus and organic matter in the sediments and subsequent reduction in the phosphorus buffering capacity.

Numerous studies have been undertaken to assess the effectiveness of phosphorus reduction to lakes. Marsden (1989) noted that although a considerable number of lakes had responded to a reduced phosphorus load as predicted, many failed to show any measurable reduction in productivity (e.g. phytoplankton biomass). The failure of these lakes to respond was primarily attributed to trophic status. In highly eutrophic lakes, phosphorus releases from sediments compensated for any reduction in external load. Furthermore it was suggested that in order to achieve significant improvements in the condition of eutrophic lakes very large reductions in external loading would be required. For example, in lakes with average annual total phosphorus concentrations of more than 100 µg/L, few improvements were recorded unless external loading was reduced by greater than 60%, whereas only moderate reductions were required in lakes with lower total phosphorus concentrations (Marsden 1989).

In mildly enriched lakes, (e.g. Lake Mjøsa (Norway), Lake Vättern (Sweden)), recovery following a reduced external load was found to be rapid. Alternatively, recovery of lakes with a long history of enrichment, such as Lake Vesijärvi (Finland) was slow, due to the ongoing internal supply of phosphorus from the sediments (Marsden, 1989).

The higher dissolved oxygen concentrations and flushing rates experienced by lotic water bodies generally results in reduced sediment released phosphorus and organic matter recycling in rivers. Additionally, as much of the phosphorus is bound to fine particulate matter, a high proportion of the phosphorus store will be transferred from un-impounded rivers, to lakes and reservoirs during high flows. Therefore, rivers would generally respond much more quickly to a reduced external phosphorus load than lakes.

It would be expected that Northern European rivers and lakes (e.g. Norway, Sweden, Finland) would respond more quickly to a reduced external phosphorus load, due to the low phosphorus concentrations in these water bodies. Conversely, those countries with a high proportion of water bodies showing elevated phosphorus concentrations (e.g. Bulgaria, Netherlands, United Kingdom etc) would be expected to respond slowly to reduced loads. However, general conclusions should not be drawn without first undertaking a thorough

review of the characteristics of each waterbody and its catchment, including historical loading data, flushing rates, sediment characteristics, morphology and present and historical catchment land use.

1.4 Project aim

The broad aim of this study is to determine the environmental and financial costs and benefits associated with substituting phosphorus in detergents with alternative builders. The study investigates the effect of banning the use of detergent phosphates on the eutrophication of surface water bodies in the EU.

The study provides an evaluation of the impact of a phosphorus ban, when implemented individually, or in combination with other practices, such as improvement to wastewater treatment. Furthermore, the study considers the cost-effectiveness of substituting phosphorus with a number of alternative detergent builders and how these may be applied in practice throughout the EU.

The subject is complex and has been the focus of a number of studies. This is compounded by the fact that any changes to phosphorus use in detergents impacts on the commercial interests of manufacturers.

This study has been undertaken without regard for commercial interests and is intended to provide a technical overview of the impacts of banning the use of phosphorus in detergents in the EU. The study has been undertaken assuming the full implementation of the UWWTD.

1.5 Project Objectives

The specific objectives of the study are to:

1. Compile all information on the legislative and voluntary measures undertaken in industrialised countries to reduce and/or ban the contents of phosphates or phosphate substitutes in detergents, and to evaluate the consequences of these measures;
2. Describe the impact on the aquatic ecosystems, particularly the risk of eutrophication, from the use of phosphorus based detergents and evaluate the relative contribution of this impact in relation to other sources (e.g. agricultural and industrial activities), and given the application of the UWWT Directive 91/271/EEC;
3. Assess the environmental and economic costs/benefits (including sludge production and disposal and recovery/use) of removal of the detergent based phosphate load in urban waste water treatment plants and compare this with the use of alternative detergent builders;
4. Provide recommendations as to the most cost effective measures to improve the present situation, with particular reference to identification of alternative detergent builders; and,
5. Describe the extraction, transport, handling and production of phosphate and alternative ingredients from the raw material to the final product as used by the detergent industry.

2. DETERGENT BUILDERS AND DETERGENT USE

2.1 Constituents of detergents

Household detergents can be classified by their use: in washing machines (laundry), fabric conditioners, for washing up at the sink, and in dishwashers. Laundry and dishwasher detergents may contain phosphorus, present as sodium tripolyphosphate (STPP). Fabric conditioners and washing up liquids used in Europe do not.

Laundry and dishwasher detergents include a wide range of compounds. The main components are builders, surfactants and stain removal agents.

- Builders develop optimum water conditions for operation of the surfactants, by deactivating hard water minerals.
- Surfactants solubilise dirt by attachment to dirt and attraction into water. They may be anionic, cationic, non-ionic or amphoteric. Anionic surfactants have the largest part of the market (>60%). They include dodecyl benzene sulphonate and linear alkyl benzene sulphonate (LAS).
- Stain removers (bleaches and enzymes) oxidise or degrade substances to decolourise them and enable removal.

Other ingredients include alkali, bleach activators, anti-redeposition agents, fluorescent agents and perfumes.

2.1.1 Detergent Builders

Builders are required to:

- Reduce water hardness (from calcium and magnesium ions which reduce surfactant efficiency and encrust fabric surfaces);
- Create and stabilise alkalinity providing conditions for optimal soil removal;
- Facilitate solubilisation of all detergent components;
- Aid dispersion of dirt and help to prevent its re-deposition;
- Maintain powder flows during manufacturing and consumer dispensing;
- Adsorb surfactants.

Builders also provide the skeleton for holding together the powder grains in a detergent. Their main uses are in laundry detergents, which account for nearly 70% of sales, followed by autodishwasher detergents (15%) and industrial detergents (15%).

Phosphates, primarily sodium tripolyphosphate (STPP), dominated the builders used from 1947 to the late 1980s. Since then STPP has been partially replaced by a combination of zeolite (mainly zeolite A) with polycarboxylic acid and sodium carbonate (Table 2.1). Zeolites are aluminosilicates. This mixture is used predominantly for washing powders (standard, compact or super-compact) or tablets.

A third system, based on citrates, is used for automatic dishwasher detergents, and liquid detergents. Citrates are less aggressive in washing delicate tableware, but are more expensive.

Table 2-1 Substances used in detergents

Name		Summary of impacts
Sodium tripolyphosphate	STPP	Contains 25% phosphorus, which (with nitrogen) is the main cause of eutrophication in rivers, lakes and coastal waters.
Zeolites (A, P, X, AX)		No measured environment effect. Increases sludge quantity. Co-built with other additives, especially PCAs.
Polycarboxylic acids	PCAs	Poorly-biodegradable, adsorb to sludge. Limited data on fate in environment; only used with zeolites.
Citrates		Chelator, more effective on magnesium than calcium ions, contributes BOD load at wastewater treatment works. Used especially for liquid detergents.
Nitrilotriacetic acid	NTA	Not used in EU, due to past fears that it may lead to increased dissolved heavy metals (chelation). These fears appear now not to have been well founded.
SUBSIDIARY COMPONENTS		
Carbonates		Softener by precipitation of calcium ions; enhances and stabilises alkalinity
Silicates		Enhances alkalinity; corrosion inhibitor
Phosphonates		Poorly biodegradable, metal ion chelator, anti-redeposition agent.
Soap		Added to reduce foaming in washing machines
Ethylenediaminetetracetic acid	EDTA	Poorly degradable. Dissolves metal ions
Carboxymethyloxysuccinate Carboxymethyltartronate	CMOS CMT	Weak chelator cf. STPP. Poor biodegradation, not trapped in primary solids; not generally used in EU.
Carboxymethylcellulose	CMC	Anti-redeposition agent, repels soil from fabrics

A fourth system, based on nitrilotriacetic acid (NTA) is not currently used in the EU as a result of concerns about toxicity and accumulation in the environment. Many of these concerns have been reduced following more recent studies.

All the builder systems now use some combination of one main component and subsidiary components to enhance the efficiency of the detergent properties (tables 2.2 and 2.3, personal communication Christopher Thornton, CEEP). There are many variants in formulations.

Table 2-2 Comparison of typical P based and P free Laundry Detergent Formulations (Conventional Powders)

%	P-Based	P-Free
sodium tripolyphosphate (STPP)	20-25	0
zeolite	0	25
polycarboxylates (PCAs)	0	4
organic phosphonates	0 to 0.2	0.4
sodium silicate	6	4
sodium carbonate	5	15
surfactants	12	15
sodium perborate	14	18
activator	0 to 2	2.5
sodium sulphate	1 to 24	9
enzymes	1	0.5
antiredeposition agents	0.2	1
optical brightening agents	0.2	0.2
perfume ¹	10	0.2
water		5

Note 1. Perfumes are not essential to the effectiveness of detergents. Their content is variable.

Table 2-3 Typical Laundry Detergent Formulations (Compact Powders)

%	P-Based	P-Free
sodium tripolyphosphate (STPP)	50	0
zeolite	0	20-30
polycarboxylates (PCAs)	0	5
organic phosphonates	0	0.2
sodium silicate	5	4
sodium carbonate	4	15-20
surfactants	14	15
sodium perborate	10	13
activator	3	5
sodium sulphate	4	5
enzymes	0.8	0.8
antiredeposition agents	1	1
optical brightening agents	0.3	0.3
perfume	0.2	0.2
water	8	5

* Monohydrated perborate is used in compacts. This is significantly more powerful bleach than the tetrahydrated perborate used in conventionals.

The precise constituents of detergents can vary between brands. While manufacturers are generally reluctant to reveal the precise constituents, an indication is given on every packet that is sold, and some data from Rhone-Poulenc is summarised in table 2.4. Three examples each of conventional and concentrated powders, both STPP based and Zeolite based, are shown. While the conventional powders are all similar, the concentrated powders vary.

Table 2-4 Constituents of some detergents

	STPP based	Zeolite based
Conventional powder	3 examples, all with STPP 15-30%, PCAs <5%.	3 examples, all with Zeolites 15-30%, PCAs <5%
Concentrated powder	STPP 5-15% STPP >30%, PCAs <5% STPP 30%, carbonates & silicates 13%	Zeolites 15-30%, PCAs <5% Zeolites 15-30% Percarbonates 15-30%

On the basis of this information, the high STPP concentration shown in table 2.3 represents an upper bound. It is assumed in the LCA calculations that both conventional and concentrated powders contain 25% of STPP or Zeolite A.

Effectiveness

Tests of the cleaning efficiency of laundry detergents have been carried out (Which? 1999, Test Achats 2000, Que Choisir 1999, Wilson et al 1994). STPP based detergents have been found by these tests to be preferred to those based on zeolite, while both types performed acceptably.

Claims have been made by trade organisations for the superior effectiveness of both STPP and Zeolite A (see for example publications by CEEP, many in the Scope newsletter, and by Zeodet. However the widespread availability and use of zeolite as a builder in countries where there is no statutory ban on STPP (such as France, Greece, UK and Scandinavia, table 2.5), shows that their effectiveness and price is widely acceptable.

Environmental impacts

Phosphorus has two main impacts:

- As a nutrient in treated effluent that can contribute to eutrophication,
- In sludge, where it contributes to the quantity and is partly available to plants.

Zeolite A is an inert, insoluble alumino-silicate (Morse et al 1994). It therefore contributes to the suspended solids concentration, and therefore to the sludge quantity. Rough calculations suggest that zeolite and polycarboxylates might comprise up to 10% of sludge dry solids if all household laundry detergents were zeolite based.

Given phosphorus removal in sewage treatment, there is probably no major difference between STPP and Zeolite A as detergent builders in terms of the quantity of sludge generated (see section 7 and appendix F).

Zeolite A has an affinity for heavy metals. The evidence on its effect on the fate of heavy metals in wastewater treatment is not conclusive, but it is believed to improve sludge settleability. There appears to be no reason to fear toxic effects (Morse et al 1994). However Morse et al do point out that if wastewater treatment is inadequate and metal levels in sediment are high, there is the risk that the hydrolysis of Zeolite A could re-release metals in soluble form. See Appendix G for more details. Other substances in sludge could contribute to the same effect.

Polycarboxylates (PCAs) are a family of synthetic polymers. Biodegradation in biological wastewater treatment is believed to be low (<20%, Morse et al 1994), and 90% or more of PCAs are believed to leave biological sewage treatment with the sludge. They have no impact on the treatment processes but may help to mobilise metals.

Because PCAs are a mixture of compounds, it has not been possible to trace their fate in the environment. As with Zeolite A, there appears to be no reason to fear toxic effects.

2.2 Types of detergent

Household laundry detergents have traditionally been powders. Compact detergents were launched in Europe in 1989, and now account for half the market. They are particularly widely used in Holland, Germany and Austria. Recently liquid gels and tablets have been introduced (AISE web site).

The German Umweltbundesamt (UBA) provides guidance on its web site on the most environmentally friendly ways to use detergents. Detergent types – all P free in Germany - are ranked in order from most friendly to least friendly, in terms of the quantities of surfactants and other chemicals discharged:

Component based system	Individual components are added separately. Most friendly in theory, but too complicated in practice
Powder for coloureds	Contain no bleach or brightener
Compact detergents & tablets	
Traditional washing powders	
Least friendly: liquid gels	

2.3 Current detergent use in Europe

2.3.1 **Builder type**

Current detergent and STPP use in Europe has been estimated from the total spending on detergents, price, and STPP content ([table 2.5](#)). The total consumption of STPP in the EU and the 3 Accession States is estimated from the percentages by country to be nearly 300000 tonnes/year. This is less than the value of 400000 tonnes/year stated by CEFIC (2000); the difference can partly be accounted for by noting that the larger CEFIC figure includes Bulgaria and Romania, where 95% of household detergents are STPP based.

It is clear from [table 2.5](#) that STPP use is relatively high in 7 (possibly 8) countries:

- France
- Greece
- Portugal
- Spain
- UK
- Hungary
- Poland
- possibly the Czech Republic.

In the rest of the EU it is either effectively zero, or low. These member states can be divided into two groups: those with a high degree of phosphorus removal in sewage treatment – Austria, Denmark, Finland, Germany, Luxembourg, Netherlands, Sweden – and those where the standard needs to be raised – Belgium, Ireland and Italy.

2.3.2 Quantity

The average consumption of laundry detergents in both the EU member and the accession states is just over 7 kg/person/year. Consumption of dishwasher detergents is 1.6 kg/person/year in the EU member states, but much lower in the accession states (0.12 kg/person/year).

The AISE is committed to reducing detergent consumption through its voluntary Code of Good Environmental Practice, in particular to a 10% reduction in consumption by 2001, compared to 1996.

Table 2-5 Estimated detergent consumption in Europe with current legislation

Country	Population (2000)	Detergent use (1998)		Detergent with STPP builder (CEFIC 2000)	STPP consumption (note 1)		% reduction 1985 to 2000
		Laundry	Automatic dishwasher		kilotonnes	kg/hd	
	millions	kilotonnes	kilotonnes	%	kilotonnes	kg/hd	
Austria	8.1	59	13	0	0	0.0	100%
Belgium	10.2	78	15	0 ²	0	0.0	100%
Denmark	5.3	31	10	20	2	0.4	90%
Finland	5.1	32	7	10	1	0.2	95%
France	58.4	450	168	50	74	1.3	60%
Germany	81.9	490	158	0	0	0.0	100%
Greece	10.5	62	9	50	9	0.8	66%
Ireland	3.6	41	3	0 ²	0	0.0	100%
Italy	57.3	415	36	0	0	0.0	100%
Luxembourg	0.4	no data	no data	no data	no data	no data	no data
Netherlands	15.5	100	21	0	0	0.0	100%
Portugal	9.9	43	2	70	8	0.8	50%
Spain	39.3	241	25	60	38	1.0	65%
Sweden	8.8	44	9	15	2	0.2	90%
UK	58.8	573	85	45	71	1.2	40%
Total EU	373.1	2659	561		205		77%
Hungary	10.2	40	1	70	7	0.7	50%
Czech Rep.	10.3	17 ³	0	65	3?	0.3?	?
Poland	38.6	372	6	85	77	2.0	15%
Total Acc'n States	59.1	429	7		87		
Total	432.2	3088	568		292		

Note 1. Assumes that detergents that use STPP as a builder contain 24% STPP, and therefore 6% phosphorus. Overall use may be under-estimated.

Note 2. These values are set to zero, assuming implementation of recent measures to control phosphorus in detergents.

Note 3. This figure is low on a per capita basis. Either it is incorrect, or it may be higher now & in the near future.

3. CASE STUDIES OF ACTIONS TAKEN TO LIMIT OR BAN PHOSPHATES IN DETERGENTS

3.1 Review of action to date

Voluntary and legislative measures to limit phosphorus concentrations in detergents, or to ban it, are summarised in **table 3.1**. To the best of WRc's knowledge, the information on legislation and voluntary agreements is complete for all the countries shown. The third column contains summaries from the case studies (see below) but is not complete for the other countries.

Detailed case studies for six EU member states, Switzerland and the USA are given below. These trace the laws and voluntary agreements on detergents that have been made, improvements in sewage treatment, and some of the achievements in terms of reduced phosphorus inputs to surface waters and observed eutrophication. The countries represent different situations and experiences:

- The Rhine is the major river system for Germany and the Netherlands. In both countries laundry detergents are now zeolite based, and sewage treatment removes phosphorus to a large extent.
- In Italy and Belgium (Wallonia) laundry detergents are now effectively all zeolite based. However progress in improving sewage treatment has generally been slow.
- In France, a significant proportion of laundry detergents currently used are STPP based.
- Hungary is an example of an accession state, and part of the Danube basin. Action to reduce phosphorus inputs is at an early stage.
- Switzerland and the USA are non EU states where successful action to reduce eutrophication has been taken.

Table 3-1 Legislative and Voluntary Frameworks for Phosphates in Detergents

Country	Legislative	Voluntary agreements	Other
European Union			
Austria	Actions to reduce quantities of phosphates in detergents were introduced in two legislative acts. The first signed in 1985 for a maximum of 24% (NTPF) and the second in 1987 for 20% (NTPF).		
Belgium	<p>The European Commission has received a draft law (February 16 2001) from Belgium which aims to prohibit the placing on the market, whether by importation or local manufacture, of domestic textile detergents containing phosphates. The reasoning provided by the Belgian government is the protection of the environment and of public health. The draft law's objectives are as follows:</p> <p>The sale or distribution by importers, local manufacturers and retailers of domestic textile detergents which contain more than 0.5% phosphorus, Irrespective of whether this is present in the form of organic or inorganic compounds,</p> <p>The draft has not yet been enacted (February 2002)</p>	A voluntary agreement signed on 18 September 1988, between the Belgian Association of Soap Manufacturers and the Government, aimed at using 100% phosphate free detergent by 1995.	There was a sharp decrease of P detergent in favour of P-free detergent from 54% on 1 January 1989 down to <10% in 1991. However, there have been some temporary small increases in two occasions in 1994 and 1999 which have set off reactions from consumers associations and green pressure groups. The Federal Government has thus proposed the draft regulation.

Country	Legislative	Voluntary agreements	Other
Denmark	None	Recommendation for phosphorus-free detergents in areas where wastewater treatment plants do not have phosphate removal, target for 50% phosphorus free detergents by 1992.	
Finland	None	Unwritten voluntary agreements were made with detergent suppliers, resulting in a 20% reduction in the quantity of P entering sewage treatment works between 1990 and 1992 (personal communication, Finnish environment Ministry).	Finland implemented the EU Directive on labelling in 1992. Since the introduction of this law, industry has voluntarily phased-out phosphorus-containing detergents, so that the current position is one of phosphorus detergents holding only a minimal market share.
France	In 1990 the Environment Secretary defined the following measures: Tri-polyphosphate levels would be limited to 25% from 1.1.1991 and 20% from 1.7.1991 in new products; Each detergent manufacturer would sell at least one phosphate-free detergent by 1.1.1991; An economic instrument has been proposed: the Taxe Générale sur les Activités Polluantes. This appears not yet to have come into force.		There has been a continuing public debate including legal action over publicity for P free detergents in the late 1980s and early 1990s. Consumer choice has resulted in currently equal market shares for STPP and Zeolite based household laundry detergents.

Country	Legislative	Voluntary agreements	Other
Germany	The phosphate content of detergents is regulated by the "Phosphate-Höchstmengenverordnung" stipulating maximum quantities. It entered into force on 1 January 1984. The content of phosphates in detergents was reduced by 50% compared with the high-phosphate ones used before that.		Phosphate free detergents have been used in Germany since about 1986 as a result of market pressure. Zeolite is used as a phosphate substitute. The use of phosphate free-detergents was a voluntary development coming from the industry itself, encouraged by public debate on the eutrophication of the aquatic environment. Since 1986 consumers have generally decided in favour of phosphate free products and since then there has been virtually no phosphates in detergents in Germany.
Greece	None	None	
Ireland	None	The Irish Government and IDAPA (Irish Detergent Industry Association) have signed an agreement to eliminate phosphorus from almost all of their products by the end of 2002. It is estimated that IDAPA members have 90% of the market and that 8% of phosphates in Irish rivers and lakes comes from detergents.	
Italy	Decree of 30.12.81, nr.801 set a limit of 5% on the P content of household laundry detergents, and required a statement of the P content on the packet. Effective from 1.1.94 Law nr.413, 13.09.88 limited the P content of detergents to 1%. Effective from 1.1.89	"Contact programs"- Voluntary agreements. First introduced in the 1970s with the region of Emilia Romagna who negotiated agreements with the chemical industry on the phosphate content of detergents.	Eutrophication became a public issue in the 1970s, as holiday areas such as the Alpine lakes and the Adriatic coast were affected. There were a number of measures at regional and national level, culminating in the law which effectively banned STPP based household laundry detergents from 1.1.1989.

Country	Legislative	Voluntary agreements	Other
Italy	Decree of 30.12.81, nr.801 set a limit of 5% on the P content of household laundry detergents, and required a statement of the P content on the packet. Effective from 1.1.94 Law nr.413, 13.09.88 limited the P content of detergents to 1%. Effective from 1.1.99	"Contact programs"- Voluntary agreements. First introduced in the 1970s with the region of Emilia Romagna who negotiated agreements with the chemical industry on the phosphate content of detergents.	Eutrophication became a public issue in the 1970s, as holiday areas such as the Alpine lakes and the Adriatic coast were affected. There were a number of measures at regional and national level, culminating in the law which effectively banned STPP based household laundry detergents from 1.1.1999.
Luxembourg	None	None	
Netherlands		Agreements made with detergent manufacturers resulted in almost all textile washing substances becoming phosphate free in 1990. This reduced the amount of phosphate reaching sewage treatment plants by approx. 40%.	Although eutrophication had been identified as a problem in the 1970s, the Dutch government did not act until international agreement on the Rhine was obtained in 1987.
Portugal	None	None	
Spain	None	None	
Sweden		The Swedish Society for Nature Conservation's Falcon label has requirements for the phosphate concentration of a number of detergent-based products: Laundry detergents 0.75 g P/kg laundry (20% STPP) Automatic dishwasher detergents 6% phosphorus All purpose and heavy duty cleaners 5% phosphorus	
United Kingdom	None	There are existing voluntary agreements with industry, which prevent the use of EDTA and NTA as builders in domestic detergents. There are no plans to create either voluntary or mandatory bans on phosphate in detergents (as of 1991)	Suggested tax on phosphate levels in detergents in DETR report, January 1998.

Country	Legislative	Voluntary agreements	Other
Acc'n Countries			
Czech Republic		Ministry of the Environment and the Czech Soap and Detergent Product Association agreement on the gradual decreasing of the impact of laundry detergents on the environment. Members of the Association voluntarily undertake to maintain maximum level content of additives in their detergent products supplied into retail network. The maximum for phosphorus is 5.5% (w/w).	
Hungary	The revised standard for the determination of total phosphate content in detergents was released in 1987. Limitations for the phosphate content in detergents were introduced by the "Standard for Pulverous Synthetic Detergents" in 1986.	Voluntary agreement between Government and manufacturers to reduce levels and improve public awareness. Introduction of Phosphate free detergents. Introduction of the European Eco-labelling system for Detergents.	Currently >50% of household laundry detergents are STPP based. Hungary's commitment to the international Danube Convention has set demanding targets for reducing P inputs. A ban on STPP in household laundry detergents has been considered in this context.
Poland	None	None	

Country	Legislative	Voluntary agreements	Other
Other Countries			
Australia	No legislative requirements.	Technology developments led to the introduction of concentrated detergents resulting in a reduced phosphate content. A voluntary labelling scheme has been introduced combined with a community education scheme concerning nutrients in water industry.	
Japan	Some prefectures where large lakes are located ban the sale and use of synthetic household cleaners containing phosphates. However, even when not specifically prescribed by law, phosphate cleaners are subject to a variety of administrative directives from the Ministry of International Trade.	There are various voluntary industry guidelines and in practice STPP based detergents are almost never sold in Japan.	
Switzerland	The use of phosphates in detergents has been prohibited since 1986.		Eutrophication occurred in lake Geneva (and elsewhere) was first recognised in the 1950s. A series of measures were introduced to improve wastewater treatment and limit the potential of detergents to pollute the water environment.
Canada	Legislation prohibiting the sale of detergents containing more than 2.2% of phosphorus became effective on 1 January 1973		
United States	27 States have either complete or partial bans on the use of phosphate in laundry detergents. These are mainly those situated along the eastern-coast or around the Great Lakes.	As a result of the number of States introducing compulsory bans, the use of phosphorus in domestic laundry detergents was phased out in 1994.	

Table 3-2 Trends in STPP consumption

Country	Estimated STPP consumption ¹ , ktonnes pa.								% reduction
	1984	1985	1986	1987	1988	1989	1990	current ²	current / 1985
Austria	17.3	18.7	16.9	13.8	13.1	11.4	10.0	0	100%
Belgium	25.5	25.5	25.5	25.5	25.5	15.3	12.2	0	100%
Denmark	18.8	18.8	18.8	15.9	17.8	16.9	13.9	2	90%
Finland	13.2	11.8	10.7	8.7	8.3	6.7	6.3	1	95%
France	180.4	180.4	175.1	154.1	137.8	132.5	127.3	74	60%
Germany	192.5	185.9	104.0	71.3	52.4	32.8	13.1	0	100%
Greece	27.1	24.2	21.8	17.9	17.0	13.7	12.9	9	66%
Ireland	9.4	8.4	7.6	6.2	5.9	4.8	4.5	0	100%
Italy	117.5	111.8	75.7	52.2	50.5	13.2	9.7	0	100%
Luxembourg	1.1	0.9	0.9	0.7	0.7	0.5	0.5		
Netherlands	38.2	32.0	32.0	5.1	2.0	1.6	1.6	0	100%
Portugal	17.9	16.9	14.9	14.9	14.9	14.9	14.9	8	50%
Spain	113.9	111.9	101.3	96.2	97.4	88.4	92.3	38	65%
Sweden	22.8	19.9	19.9	17.9	16.4	11.6	10.9	2	90%
UK	127.6	122.3	133.4	123.4	130.5	125.2	127.6	71	40%
Total EU	923.2	889.4	758.5	623.8	590.2	489.5	457.7	205	77%
Hungary	26.3	23.5	21.2	17.4	16.5	13.3	12.5	7	50%
Czech Republic	26.7	23.8	21.5	17.6	16.7	13.5	12.7	3	?
Poland	99.8	89.1	80.5	65.8	62.5	50.6	47.5	77	15%
Total Accession States	152.8	136.4	123.2	100.8	95.7	77.4	72.7	87	
Switzerland	24.0	21.9	9.8	4.4	4.4	3.3	2.2		

Note 1. Based on per capita values from CES (1991). STPP contains 25% phosphorus.

Note 2. See [table 2.5](#). Assumes effective application of recent action in Belgium & Ireland. Values may be underestimates in some cases, in particular for the Czech Republic.

3.1.1 International Conventions

Some of the case study countries have signed international conventions, which have influenced their actions on pollution control. A summary of 4 important conventions is given here.

OSPAR

The Convention for the protection of the Marine Environment of the North East Atlantic (OSPAR Convention) entered into force on the 25th of March 1998. The Convention was established in recognition of the importance of the marine environment, and the necessity for providing co-ordinated protection for it. To achieve this it was agreed that national, regional and global action

was required to prevent and eliminate marine pollution and to achieve sustainable management of the maritime area to continue to meet the needs of present and future generations. The Convention replaces the Oslo and Paris Conventions.

The parties to the Convention are: Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Netherlands, Norway, Portugal, Spain, Sweden, UK, Luxembourg, Switzerland and the European Commission.

The Contracting Parties are required to take, individually and jointly, all possible steps to prevent and eliminate pollution of the marine environment from land-based sources, including pollution from dumping or incineration of wastes and pollution from offshore sources.

To enable assessment of the quality of the marine environment, the Contracting Parties are required to undertake and publish joint assessments of the quality status of the marine environment and of its development. These assessments must include both an evaluation of the effectiveness of the measures (taken and planned) and the identification of priorities for action. The Convention focuses on the 'precautionary' and 'polluter' pays principles and has the requirements that Members adopt programmes (joint or complementary) of scientific or technical research and harmonise policies and strategies.

Danube

The Danube Convention entered into force on the 29th June 1994. The Convention was established due to concern over short and long term threats of adverse effects of changes in watercourses within the Danube River Basins on the environment, economics and well being of Danubian States. Danubian States include Sovereign States that share a considerable part (>2,000km²) of the hydrological catchment area of the Danube River. The Convention was signed and ratified by six countries (Austria, Croatia, Czech Republic, Germany, Hungary and Romania) by September 1997.

The objectives of the Convention are to achieve sustainable and equitable water management including the conservation, improvement and rational use of surface and groundwater in the catchment. Member States must make efforts to control hazards from accidents involving hazardous substances, floods and ice hazards and contribute to reduced pollutant loads to the Black Sea. The Convention requires, at a minimum, to maintain and improve the current environmental and water quality conditions of the Danube River and waters within its catchment.

To achieve the objectives, Member States are required to strengthen, harmonise and co-ordinate measures at a national and international level aimed at sustainable development and environmental protection in particular to ensure sustainable use of water resources (municipal, industrial, agricultural purposes), conservation and restoration of ecosystems and public health.

Rhine Action Plan and Convention

Due to the large number of industries located on the banks of the river, as well as the potentially dangerous cargoes carried by ships on the river, chemical pollution was reaching high levels by the late 1980's. This culminated in the disastrous fire in the Schweizerhalle pesticide chemical

plant in 1986, which led to serious pollution of the Rhine. After this event the International Commission for the Protection of the Rhine (ICPR) drew up a Rhine Action Plan Against Chemical Pollution, which was adopted by the European Community in 1987. The RAP imposes a strict regulating regime on industries alongside the river, including limitations on the application (amount and type) of agricultural chemicals.

The Convention on the Protection of the Rhine entered into force on April 12, 1999. The objectives of the Convention are to promote sustainable development of the Rhine ecosystem which applies to surface and groundwater, ecosystems that interact with the Rhine and the catchment area. The requirements of the Member States include:

- Maintain and improve the quality and function of the Rhine's waters, including the quality of suspended matter, sediments and ground water
- Protect populations of organisms and species diversity and reducing contamination by noxious substances in organisms;
- Conserve, improve and restore natural habitats to improve the ecological health of the river system (including in the water, substrate, banks and adjacent areas); and,
- Take account of ecological requirements when implementing technical measures to develop the waterway, e.g. for flood protection, shipping or the use of hydroelectric power.

The Contracted Parties to the Convention are Germany, France, Luxembourg, the Netherlands, Switzerland and the European Community.

The Helsinki Convention

The Baltic Sea has a history of eutrophication resulting from runoff from coastal areas, exchange of water with the North Sea, atmospheric depositions and human activities at sea. This has resulted in severe hypolimnetic oxygen depletion, production of hydrogen sulphide and algal proliferation in many areas. The Baltic Sea is almost totally enclosed by land and only connected to the North Sea by narrow shallow straits around Denmark and Sweden. This limited exchange capacity with the North Sea (residence time of around 25-30 years) has compounded the eutrophication problem. The North Sea is an important source of oxygen to the Baltic and in 1993, masses of high salinity water entering from the North Sea ended a 16 year period of stagnation. This was the first time since 1977 that the Baltic was free from hydrogen sulphide.

Despite substantial improvements in wastewater treatment, eutrophication was still a problem and in 1992 the Convention on the Protection of the Marine Environment of the Baltic Sea Area (The Helsinki Convention) was established, which entered into force on 17 January 2000. The Convention was implemented in recognition of the hydrographic and ecological values of the Baltic Sea Area and the sensitivity of its living resources to environmental changes.

The objectives of HELCOM are to prevent and eliminate pollution in order to promote the ecological restoration of the Baltic Sea Area and the preservation of its ecological balance. To

achieve these objectives, Member States are required to take all possible steps to prevent and eliminate:

- Pollution of the marine environment of the Baltic Sea Area originating from land-based sources;
- Pollution of the marine environment of the Baltic Sea Area caused by harmful substances

The present contracting parties to HELCOM are Denmark, Estonia, European Community, Finland, Germany, Latvia, Lithuania, Poland, Russia and Sweden.

3.2 Walloon Region of Belgium

The Walloon region occupies the western part of Belgium. The total population is 3.34 million (Statesman's Yearbook, 2001), and the main towns are Charleroi, Liège and Namur.

3.2.1 Legislative background

Product legislation and policy

A voluntary agreement was signed on the 12 of September 1988 between the Belgian Association of Soap Manufacturers and the Belgian Federal Government aimed at promoting P-free domestic laundry detergents in Belgium. The defined objectives of this agreement were for example:

placing on the market at least one P-free detergent within 6 months from the beginning of this agreement and to maintain the sale during the time span of this agreement;

P-free products will be placed in sufficient quantities, of comparable quality and price than conventional detergents;

- Providing composition on packaging of laundry detergent;
- Supplying every 6 months information on market share of P-free detergents (liquid and powder).

The results of these market surveys are presented in Figure 3.1 below. The figure shows clearly a sharp decrease of P detergent in favour of P-free detergent from 54% on 1 January 1989 down to 2.6% in September 2001. However, there have been some temporary small increases in two occasions in 1994 and 1999 which have set off reactions from consumers associations and green pressure groups. The Federal Government has thus proposed a draft regulation to ban the placing on the market of domestic laundry detergents containing more than 0.5% P. The original deadlines were 1 January 2002 for banning of P detergents for distributors and 1 July 2002 for banning of P detergents for the public. The proposed legislation is still pending (in April 2002) after comments from the European Commission requesting some amendments. In the meantime the industry is still working according to the voluntary agreement.

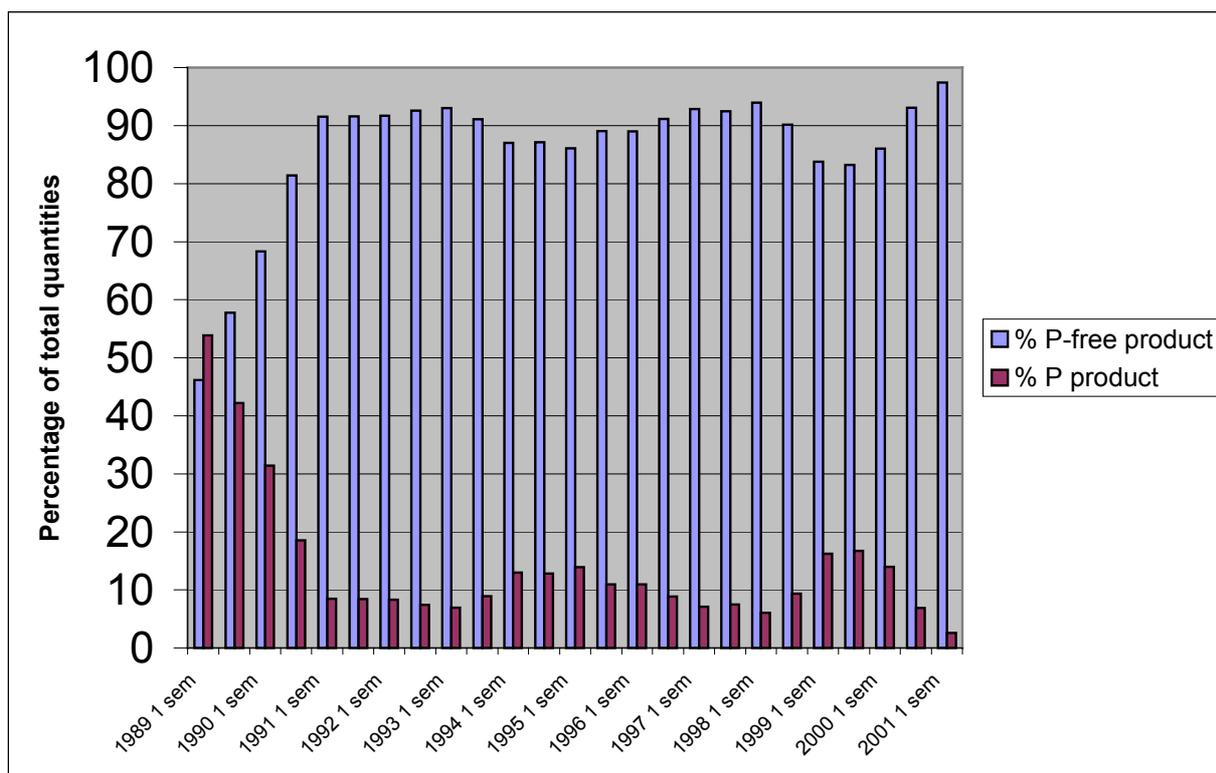


Figure 3.1 Trends in domestic P-free laundry detergent in Belgium (DETIC, pers com 2001)

Effluent regulations

Under the UWWT Directive 91/271/EC, the whole of Walloon Region has been designated as sensitive area requiring the upgrade and construction of WWTP with nutrient removal for agglomerations > 10,000 PE and compliance with N and P emission limits as defined in the Walloon Order of 25 of February 1999 (MB 27.03.1999) as amended by Order of 8 of February 2001 (MB 17.02.2001). The majority of the WWTP plants operating in the Walloon region are of size below 10,000 PE (Table 3.3). Currently, of the 300 existing WWTPs, only 16 including 4 under construction have P removal (Table 3.4).; these represent less than 10% of the total equivalent population. The majority of these works rely on biological treatment with post chemical precipitation. Some large agglomerations are in the process of upgrading and building WWTPs which will be all fitted with P removal.

There are P emission limit values for specific industrial sectors (i.e. asbestos-cement; textile, paper, pig farm, refinery, etc). New norms will be implemented under the integrated permit ('*Permis unique*')

Table 3-3 Type of WWTP plants in the Walloon Region

Class	<2,000 PE	2,000-9,999 PE	10,000-49,999	>50,000
Escaut	39	16	8	5
Meuse	130	61	17	1
Rhin	12	1	1	0
Seine	1	0	0	0
Total	182	78	26	6

Table 3-4 List of WWTP in the Walloon Region with P removal

Localisation	Capacity (pe)
Arlon	30,000
Baelen Membach	24,600
Bertrix	8,500
Butgenbach	3,200
Ciney	16,000
Durbuy Bomal*	11,300
Enghien Marq	15,000
Genappe Ways	9,800
Hannut Avernas-le-Baudoin*	9,200
La Louviere Saint Vaast	19,000
La Louviere Trivieres*	36,800
Peruwelz*	14,000
Plombieres La Geule	24,750
Tournai Froyennes	50,000
Waimes Robertville	800
Waterloo	20,000
Total	292,950

Note:*= under construction

3.2.2 Impact on industry

There are 3 large detergent producers in Belgium; Procter, Henkel and McBright. It is reported that large producers in Belgium have now shifted to P-free detergent especially since producing tablets and liquid detergents (DETIC, pers com 2001). It is reported that 90% of household detergents were zeolite-based in 1998 in Belgium (Zeodet publication). The STTP consumption in Belgium was reported to have decreased since 1988 from 25.5 ktonnes per annum (ktpa) to less than 5 ktpa in 1998.

However, P detergents are still available on the Belgian market when distributed from France and Spain. The influence of a total ban on P detergent is expected to have little impact on the main players of the industry in Belgium but will affect some smaller companies distributing P detergents (DETIC, pers com 2001).

A survey carried out in March 2000 by a consumer association (Test Achats No430) tested 16 conventional washing powders including 5 containing P and 6 tablets including 4 containing P.

The price per wash (November 1999 basis) for powder varied between 0.25 to 0.42 euros (10 to 17 BF) for conventional P free powders and 0.25 to 0.35 euros (10 to 14 BF) for P powders. The price per wash for tablets varied between 0.32 to 0.49 euros (13 to 16 BF) for P free tablets and 0.27 to 0.35 euros (11 to 14 BF) for P tablets.

There is currently no suitable substitution for P product for dish-washing detergent which usually contain between 20 and 30% P (DETIC, pers comm 2001). In Belgium, it is reported that 20 to 30% of households have a dishwasher.

3.2.3 Surface water quality

River Meuse

The Meuse rises in France, enters Wallonia at Dave upstream of Andenne, flows through Namur and Liège, and then enters the Netherlands at Visee. In Holland it flows past Maastricht, Venlo and Nijmegen.

There have been signs of eutrophication in the river Meuse from the early 80's. Algal blooms are common in spring and decrease in early summer probably due to rotifer grazing. However no clear relationship could be established between an increase in algae growth and dissolved nutrient concentration at least in the Belgian part of the river (Descy 1992, Descy 2001, pers. comm). There seems to be some relationship with increased P concentrations in the French section of the river mainly due to increased connections to sewer in the 70's of domestic effluent without N and P removal.

In 1994, an inventory was carried out by the International Meuse Commission to assess the quality of the river Meuse. The mean annual concentration of Total P in the river Meuse based on 9 sampling points along the river varied from 0.15 to 0.94 mg/l (CIPM 1997). Since 1998, The River Meuse Commission has implemented a homogenous monitoring programme across the 3 countries; concentrations are measured every 4 weeks at 17 sampling stations. The Commission reports on this information annually, and reports are available for 1998 and 1999.

Because the collection of information on a consistent basis began late, quantified trends are not available. However the 1998 and 1999 data show that total P concentrations are still high (Figure 3.2). The increase in total P between Andenne and Visee is probably due to discharges from Namur and Liège.

Chlorophyll *a* is monitored from May to September at different stations along the river. There has been a steady increase in algal biomass since 1980. Even after the introduction of P free detergent, no reduction in chlorophyll *a* concentration was seen (Descy 2001, pers. comm). The information for 1998 and 1999 is summarised in Figure 3.3; the peak values are at Agimont, further upstream than the peak P concentrations. The reason for this is not known.

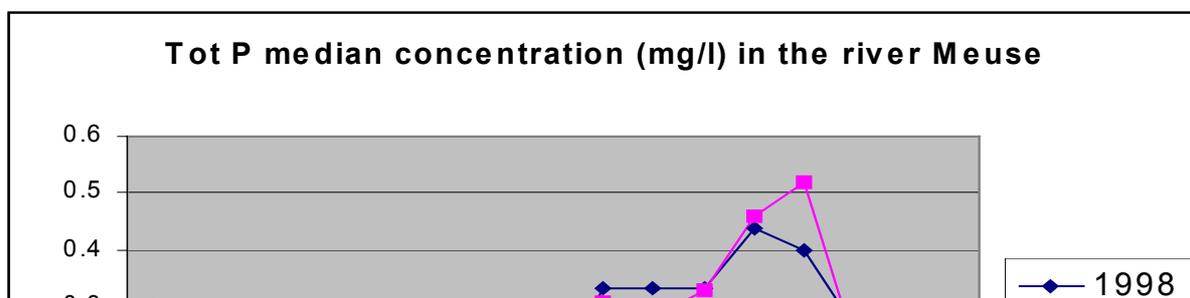


Figure 3.2 Comparison of median concentrations for Tot P, Meuse

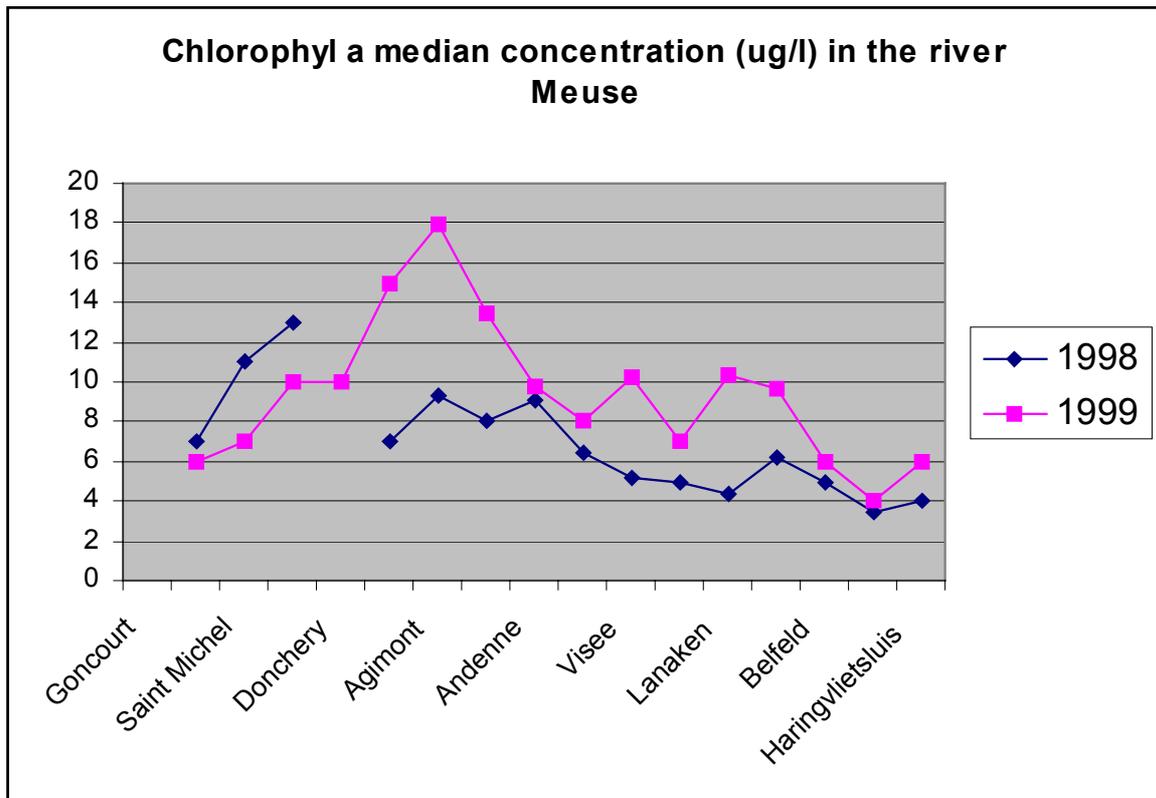


Figure 3.3 Concentration in Chlorophyl a, Meuse

River Schelde

The Schelde enters Belgium from France at Bléharies. It flows through Gent (at Zingem) and Antwerp (at Dendermonde) before entering the Westerschelde estuary.

On the river Schelde, there is also an International Commission, which has implemented a homogeneous monitoring programme on 14 stations since 1998 (Figure 3.4). The high P concentrations occur where the Schelde flows through urban Belgium. Dilution occurs in the estuary. The concentration of chlorophyll a is presented in Figure 3.5; its peak coincides with the peak P concentration.

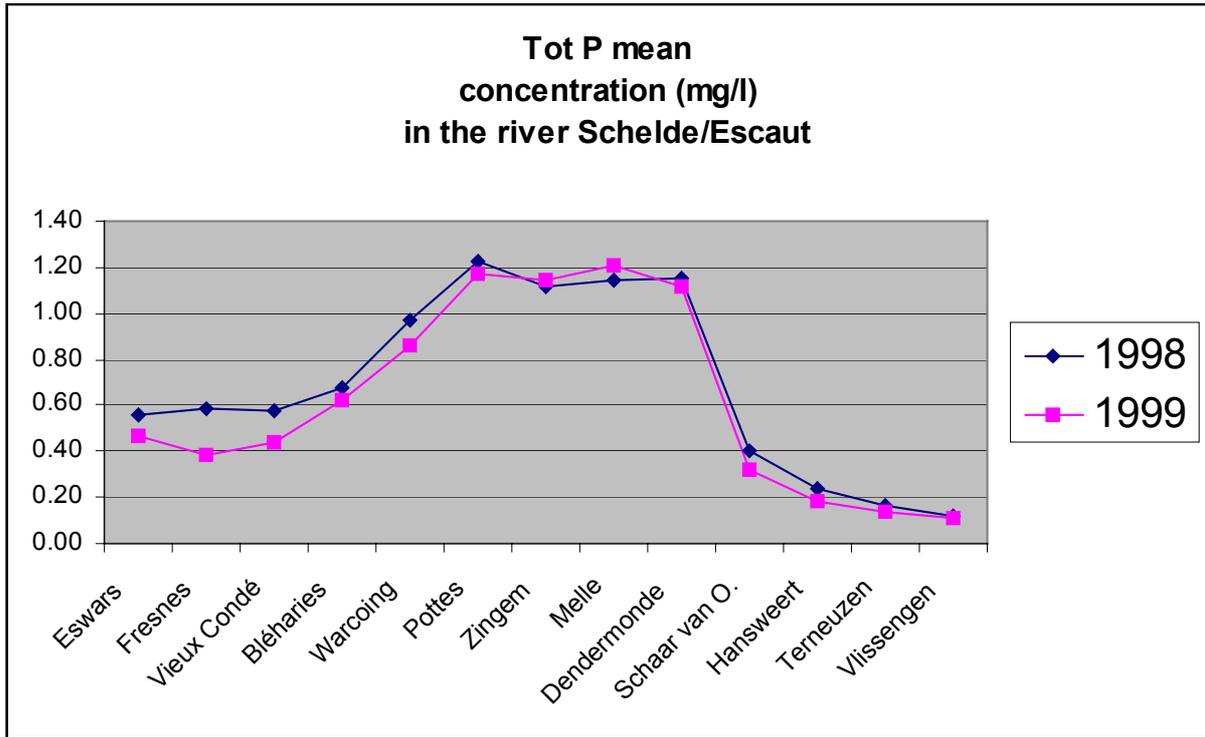


Figure 3.4 Total P concentrations, Schelde

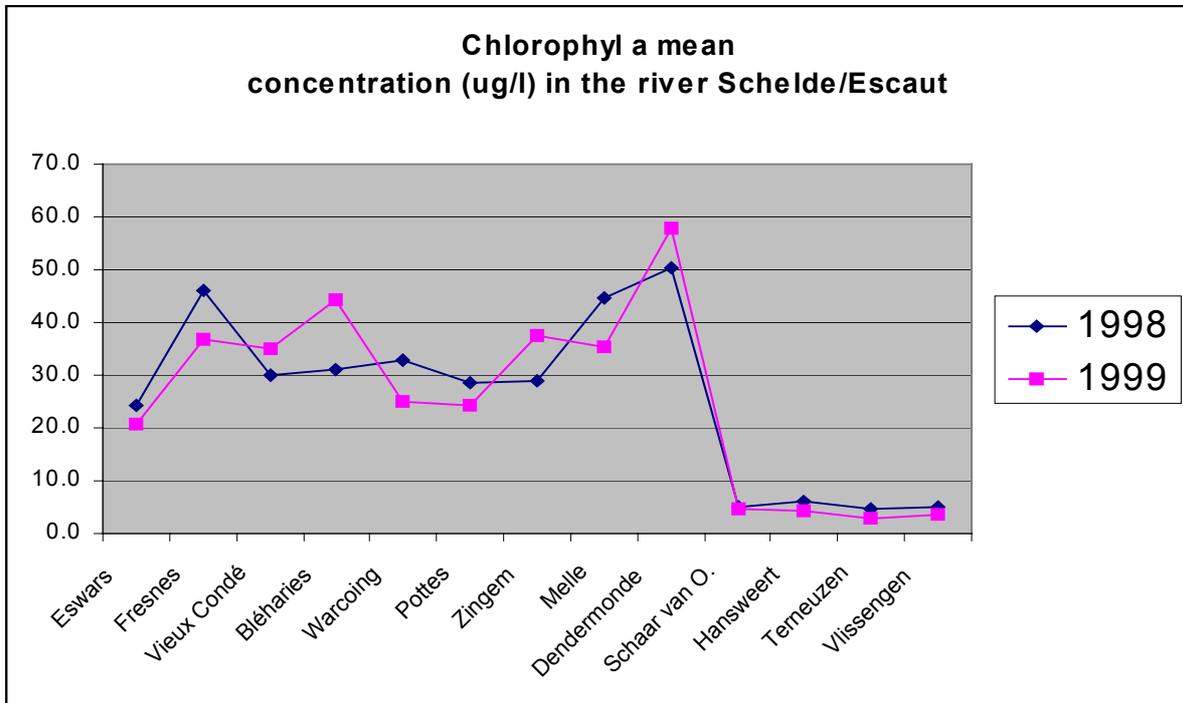


Figure 3.5 Chlorophyll a concentration, Schelde

3.2.4 Nutrient loading

The information available on nutrient loadings in different river catchments of the Walloon Region has been provided by the Water Division of General Direction for Natural Resources and Environment (DGRNE). The data were collected for the next North Sea Conference being held in Oslo in March 2002.

The nutrient load from urban waste water treatment plants is given in Table 3.5 below. It was reported that there have been very limited changes in P input into wastewater treatment plants and no trends could be drawn from 1994 to 2000. It was not possible to receive information pre 1994 to see the effect of the decline in sale of P detergents following the agreement on P-free detergents.

The load from agglomerations connected or planned to be connected to sewer in 5 to 10 years but not currently treated is given in Table 3.6. The largest proportion of P load from agglomeration is from domestic sources. For example P load from domestic source not yet connected to a sewage treatment plant amount to 1050 t per year compared with 44 t per year from industries.

The nutrient load from individual dwellings is presented Table 3.7. The nutrient load directly discharged into the environment from industrial sectors is given in Table 3.8 below. The nutrient loading from agriculture and surface run-off is given in Table 3.9 below.

Industrial loading of nutrient is relatively low compared with sewerage and sewer discharges as well as agriculture and run-off discharge (table 3.10).

Table 3-5 Treatment efficiency and nutrient loading from WWT plant (tonne per year)

Catchment	N in	N out	N eff (%)	P in	P out	P eff (%)
Schelde/Escaut	2510	1034	59	286	141	51
Meuse	2308	1208	48	364	164	55
Rhine	80	39	52	13	5	59
Seine	1	1	11	0.1	0.1	15

Source: DGRNE 2002

Table 3-6 Nutrient load from existing and future sewerage network* (tonne per year)

Catchment	N	P
Schelde/Escaut	1901	313
Meuse	4517	775
Rhine	32	5
Seine	0	0
Total	6450	1094

Source: DGRNE 2002

Table 3-7 Nutrient load from individual habitat (tonne per year)

Catchment	Tot N	Tot P
Schelde/Escaut	97	10
Meuse	261	26
Rhine	22	2
Seine	1	0
Total	381	38

Source: DGRNE 2002

Table 3-8 Nutrient load from direct discharge from industries

	Number	Tot N (t/y)	Tot P (t/y)
Catchment Schelde/Escaut			
Cookeries/refineries	2	62.14	1.06
Fertiliser industry	2	280.36	4.73
Food/drink related industry	17	74.19	6.4
Organic chemical/bio-chemical industry	13	95.34	9.19
Other sectors	30	342.14	2.63
Waste processing industry	3	20.54	0.06
Sub-total	67	874.71	24.07
Catchment Meuse			
Cookeries/refineries	2	103.83	0.2
Fertiliser industry	1	4.92	109.05
Food/drink related industry	30	103.48	22.5
Organic chemical/bio-chemical industry	10	56.16	5.63
Other sectors	85	181.02	6.14
Pulp/paper industry	3	102.45	6.41
Waste processing industry	16	73.49	1.16
Sub-total	147	625.35	150.09
Total 1999	214	1500.06	174.16

Source: DGRNE 2002

Table 3-9 Diffuse nutrient losses (tonnes per year)

Catchment	Tot N	Tot P
Schelde/Escaut	7431	338
Meuse	10446	578
Rhine	499	36
Total	18376	952

Source: DGRNE 2002

Table 3-10 Summary of P inputs to river systems

Catchment	Total P input (tonnes/year)					Total
	Treated sewage effluent	Untreated sewage	Isolated dwellings	Industries	Diffuse (runoff, animals)	
Schelde/Escaut	141	313	10	24	338	826
Meuse	164	775	26	150	578	1693
Rhine	5	5	2	0	36	48
Total	310	1093	38	174	952	2567

3.2.5 Conclusions

In Belgium, 90% of domestic laundry detergents sold have been built from Zeolite A since the introduction of a voluntary agreement in 1988. A draft regulation to ban the sale of STPP based domestic laundry detergents in Wallonia was introduced in 2001, but has not yet become law.

Phosphorus removal has been installed at a minority of sewage treatment works in Wallonia, and much sewage is still discharged untreated. Eutrophication has occurred in the river systems since 1980, and still occurs. The main sources of phosphorus are treated and untreated domestic wastewater (55%) and diffuse sources including agriculture (37%).

3.3 France

3.3.1 Voluntary agreement on detergents

- In 1990 the French Environment Minister published an official report into detergent phosphates and the environment. Following this report, the French Environment Ministry agreed with the International Soap and Detergent Association (AISD - Association des Industries des Savons et des Détergents), a series of different actions to target the problem of high phosphate concentrations within surface waters; Tri-polyphosphate levels to be limited to 25 percent (as from 1 January 1991) and 20 percent (as from 1 July 1991) in new products. The phosphates content of French detergents approximated these levels when the measure was introduced;
- Each detergent manufacturer to sell, by 1 January 1991, at least one phosphate-free detergent. This was already generally the case when the measure was introduced;
- EDTA to be limited and use of NTA to cease within France. This corresponded to the existing situation when the measure was introduced; and
- An expert committee to look into aspects of the environmental effects of detergents, both phosphate-containing and phosphate-free formulations.

In addition, manufacturers agreed to label all packaging with advice on washing and dosage within areas of differing water 'hardness'. The AISD estimated that phosphate use in detergents dropped by 16% in France in 1990 and that zeolite use increased by 64%.

It is reported that there was a 30% reduction in quantities of STPP in detergents in France between 1990 and 1996 (Ifen 1999).

3.3.2 UWWTD

The UWWT Directive was transposed into French national law in 1994, after identification of sensitive waters. The sensitive areas delimited in 1994 covered approximately one-third of the total land area of France. The national list was revised in 1999 and, as a result, there has since been a slight increase in the total surface area.

France updated its legislation by means of the Water Act No. 92-93 of 3 January 1992 (Articles 2, 10 and 35), Decree No. 94-469 of 3 June 1994 and the Ministerial Orders of 22 December 1994. Prior to this, discharges of industrial waste-water into urban waste water systems and receiving waters had been regulated, from 1976-1993, by national legislation.

3.3.3 Economic instruments

A tax targeted at phosphates in household detergents was proposed in January 2000: the General Tax on Polluting Activities (Taxe Générale sur les Activités Polluantes - TGAP). The text was however contested by the Constitutional Council and is currently being discussed in Parliament. It has been suggested that this proposal may be cancelled.

The TGAP contains several different taxes, aimed at various activities, including use of potentially polluting laundry detergents. A stated objective of the TGAP is to reduce polluting activities through an improved application of the polluter pays principle. It is also presented as having the objective of modifying consumer behaviour. The part of the tax applied to detergents would be levied on the sales price to the consumer (FMF, 2000).

The tax represents 2.35-2.85 FF for a 1 kilo standard detergent packet, which is between 2-3% of the sale price for concentrated powders and 10% for the cheaper powders (Köhler, 2001). The expected revenue for the first year was 500 million FF, out of a total of 4 billion FF for the TGAP.

An assessment of the TGAP by Köhler (2001), believes the tax to have the following positive aspects:

- As the use of phosphates in detergents is taxed, the TGAP addresses eutrophication directly;
- The TGAP is reasonably effective in political terms, addressing eutrophication whilst remaining small enough not to impose large additional costs on the consumer.

However, Köhler also believes there are a number of limitations to a national tax on phosphorus in detergents. The main limitation is that the introduction of a national tax does not account for local variations. For example, in catchments dominated by waste water impacts, a tax on STPP in

detergents is likely to contribute significantly to a reduction in surface water phosphate concentrations, while the effect will be much less in a catchment dominated by agricultural runoff. Therefore a large proportion of taxpayers will be facing extra costs with little observed gain. Measures to reduce phosphorus loads to surface waters should include a combination of reduced phosphate use in detergents, improved wastewater treatment and reduced agricultural inputs (e.g. impose a tax on agricultural P as well as detergent P).

Kohler also points out that:

- The demand for detergents is relatively inelastic, i.e. consumers will always want to wash clothes and are not likely to be very sensitive to changes in detergent prices;
- Household expenditure on detergents is a small proportion of overall household expenditure, therefore even a high rate of tax would be unlikely to cause consumers to use less detergent;

Coupled with the observation that the effect of the tax on detergent prices would be fairly minor ($\leq 10\%$), it seems probable that the tax, if introduced, would have little effect on customers' choice of detergent.

3.3.4 Public Reaction to Actions

There is widespread opinion that phosphates contribute to eutrophication of surface water bodies, the consequence of which has been a reduction in the use of STPP (sodium tripolyphosphate) in detergents (table 3.2, Köhler, 2001).

The NF Environment Label and Retour Brand are the dominant eco-labelling schemes within France, assessing product manufacturer's claims of producing environmentally 'friendly' products, including detergents. While such labelling of phosphate-free detergents does not by itself tackle the problem of phosphorus-related eutrophication, it is widely believed that eco-labelling dissuades manufacturers from misleading the general public.

3.3.5 Waste water treatment

Definitive up to date information is currently being collated by IFEN, but is not yet available. However, data provided by IFEN and from other sources (EU 2001, OECD 1997 and Eurostat 1995) provide an overall picture.

- Sewerage. In France 80% of the population is connected to a sewer; about 10% rely on individual treatment and 10% discharge directly into the environment.
- Sewage treatment. About 77% of households are connected to a sewage treatment plant, i.e. nearly all of those connected to a sewer. There are around 15,000 urban waste water treatment plants in all.
- Agglomeration size. It is estimated that 62% of the population is in agglomerations of more than 10,000 pe. The remaining 15% of the population whose sewage is treated are in small agglomerations (< 10000 pe).

- Nutrient removal. Waste-water treatment plants are being progressively equipped for nitrogen and phosphorus removal. In 1995, there were approximately 200 waste-water treatment plants with the capacity to treat phosphorus. This number compares with just 22 equipped plants in 1986. The EU reported (EU 2001) that of 61 large cities, 6 have full tertiary treatment, 28 have full secondary treatment and 17 (including Paris) have primary or incomplete secondary treatment.
- Different estimates of the amount of phosphorus removed by sewage treatment have been made. In 1996, a study of the performance of six different sewage works was published, detailing the removal rate for different detergent components (Cemagref, Groupement d'Anthony, 1996). Their results indicated that the two sewage works using activated sludge treatment were achieving over 50% phosphorus removal. A lower efficiency rate was reported for the waste-water treatment plants in agglomerations above 10,000 pe, around 23% in 1996 (Ifen 2000).
- Estimates for the current situation and the future full implementation of the UWWT Directive have been made using reasonable assumptions (table 3.11). These show that, while a considerable reduction is expected from implementing the directive fully, a considerable proportion will still be discharged to the water environment – some of it in sensitive areas from small agglomerations.

Table 3-11 Estimated proportions of total P removed in sewage treatment: France

Treatment level	% Removal of total P	% Wastewater	
		Now	After UWWTD implemented
Local	50%	20%	20%
None	0%	3%	0%
Primary	20%	20%	0%
Secondary	40%	47%	30%
Full tertiary	85%	10%	50%
Overall		41%	65%

Note. The % receiving full tertiary treatment after implementation of the Directive has been estimated from the EU view of sensitive areas, and the proportion of the population in agglomerations >10000pe.

3.3.6 Surface water quality

Lac du Bourget (Jacquet et al, INRA, personal communication, 2002)

Lake Bourget is the largest French natural lake. It is located in the Savoie Alps at an altitude of 231m. Its mean depth is 80m, and maximum depth 145m. Its area is 42 km².

The quantities of phosphorus entering the lake have been reduced from 300 tonnes in 1974 to 94 tonnes in 1996. The P concentration in the lake has reduced in proportion. M. Jacquet states that

phosphorus present in the sediment could potentially be released at up to 60 tonnes/year, if anoxic conditions were to occur.

Transparency and chlorophyll a data indicate that the quantity of phytoplankton remains high. The nature of the community changed in 1996 from filamentous forms to predominantly the toxic cyanobacterium *Planktothrix rubescens*. The lake is still meso- to eutrophic (OECD criteria).

	1974	1982	1987	1996	2001
Total P entering the lake (tonnes/yr)	300	150	135	94	
Average P concentration (ug/l as P) ¹					
– Orthophosphate	~100		~70		~18
– Total P					~26
N/P ratio in early spring	<~6		<~15		<~28
Average transparency (m)		~4		~6	6.2
Chlorophyll a (ug/l)			~9.2	6.2	7.2

Note 1. Through a water column in the middle of the lake

ETC river data

Data reported by countries to the European Topic Centre on Water provides a record of changing total phosphorus concentrations within the four main rivers in France, the Rhone, the Seine, the Garonne and the Loire. Figure 3.6 shows as an example measured concentrations at representative stations over the period 1986 to 1998. Some reduction in total P can be seen for the Loire and the Seine, but the concentrations of phosphorus are still high in 1998.

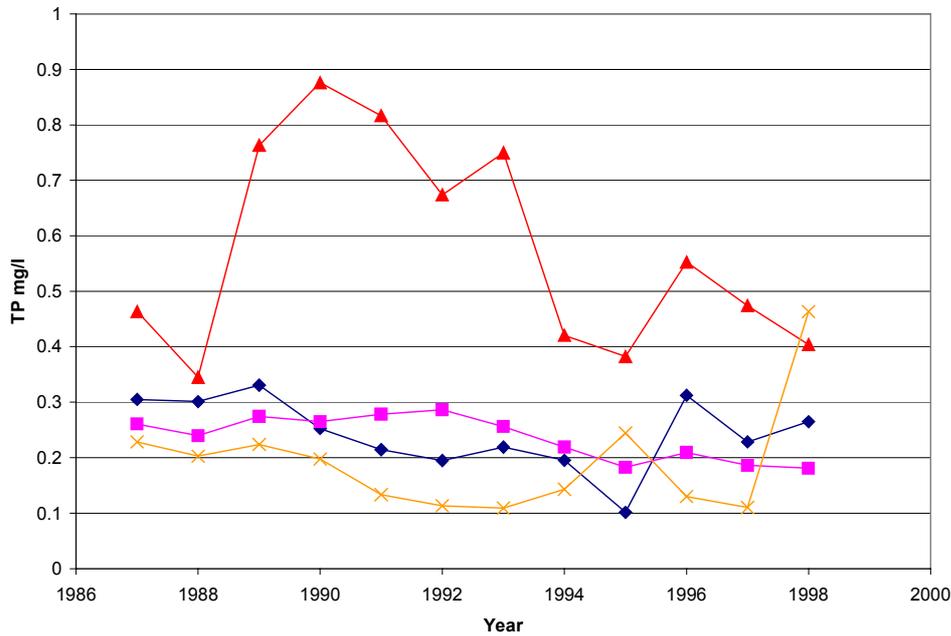


Figure 3.6 Total P concentrations in 4 French rivers

Loire catchment

Eutrophication has been detected in the Loire catchment since the first pollution inventory in 1971. In 1999, 45% of its rivers were classed eutrophic (Ifen 1999; eutrophic index E3 to E5, *moyenne, forte* and *très forte*). The average for the whole of France is 29%, with rivers of all sizes being affected.

Eutrophication has remained at constant level since 1980, in spite of improved sewage treatment and reduced P inputs to rivers. This is believed to be due to the reduction in discharge of toxic elements, which were previously limiting algae growth (Ifen 1999).

Lower Seine

The total P concentration in the lower Seine has fallen since 1993, but was still high in 1998 at 0.4 mg/l (fig. 3.6). This is consistent with information from Ifremer who state that the quality of the Seine from above Rouen to the sea is polluted (class 3); until 1997 some sections below Rouen were classed as highly polluted (class 4).

The flux of total P to the sea has reduced, to a large extent because industrial inputs have fallen from 105 t/day in 1974 to 4.3 t/day in 1998. The total flux in 1998 was 42.3 t/day. The main contribution was domestic sewage, from Rouen and urban areas upstream including Paris.

The flux above Rouen has also reduced, from an average of ca. 60 t/day between 1974 and 1989 to 40 t/day in 1998. It is likely that some of this reduction is due to the reduced use of detergents containing STPP.

In spite of these improvements, eutrophication remains a problem. Ifremer cite in particular the tributaries of the lower Seine, such as the Iton, and coastal rivers such as the Charentonne. Total P concentrations are normally higher than 0.3 mg/l, high concentrations of chlorophyll indicate the presence of phytoplankton between April and October. Ifremer (2001) note that phosphorus is the limiting element for eutrophication in the coastal waters of the Baie de Seine, particularly in the Spring.

Comité Environnement Détergents study

In 1999, the French Comité Environnement Détergents (CED) presented the results of several years of research into the potential risk to the environment of detergents (CED, 1999). The research programme, covering a range of studies, involved over 50 experts and scientists and a steering committee including representatives of industry, regulators, environmental and consumer associations. The work included:

- A Comparison of concentrations of different detergent components likely to be found in rivers (using a detailed model of 800 river reaches) with concentrations liable to have an environmental impact; and,
- Measurement of the removal rate achieved for different detergent components in six sewage works of different configurations and types;

A comparison between model-predicted concentrations was undertaken of various detergent components in 800 river reaches at risk of environmental impact. The model took into account the existing detergent use, based on national averages and sewage treatment (connection rate, type of sewage works, operating efficiency).

Thirteen separate components, including phosphates and zeolite were modelled. For each component, Predicted Environmental Concentrations (PEC) were compared with calculated Probable No Effect Concentrations (PNEC). The study found that for zeolites, PEC did not exceed PNEC on any occasion. Two potential impacts of phosphorus (as STPP) were considered:

- Through its toxicity. In no cases did the PEC exceed the PNEC.
- Through its contribution to eutrophication. Here the committee acknowledged the complexity of the issue and the need to consider the nature of each receiving water. No conclusions were drawn on this issue.

While the study contains much detail on the constituents of detergents that are a potential risk to the environment, it is not relevant to the issue of eutrophication.

Redon river

A report presented by the Comité Environnement Détergents (CED), looking at phosphorus inputs into Lake Geneva and into the river Redon, a small tributary of the lake with a rural catchment, concluded that, detergents contributed only 7% of total phosphorus inputs to Lake Geneva in 1986. It is not clear whether this figure is for all inputs to Lake Geneva, Swiss as well as French. This assumed that 50% of municipal and industrial waste water phosphorus came from detergents, which would correspond to universal use of detergents built from STPP. These results are in contrast to a detailed study that was carried out of all the point sources along the Redon river, which found 16-20% of phosphorus in the Redon River came from detergents. The CED considered this estimate to be unrepresentatively high because only one fifth of the Redon catchment's population were connected to sewage treatment at the time. Furthermore, they concluded that a change in use to P-free detergents in the basin would not lead to measurable differences in phosphorus concentrations flowing from the river into Lake Geneva.

Vilaine River

A study by the Institute Scientifique at Technique de l'Environnement (Moreau *et al*, 1998) investigated total phosphorus loads compared with export in the water of the Viliane River (above Rennes) over a one year period during 1994-1995.

The catchment comprises 902 km², of which 78% is used for agriculture. The main town is Vitré, with a population of 15000; the total population equivalent is 60000.

River water was sampled for flow rate and nutrient concentrations at 20 different sampling sites on the Vilaine and on tributaries within the upper Vilaine catchment, enabling the contribution of subcatchments to be studied. Sampling was carried out every 2 weeks during periods of low flow, and more frequently during high flows. The river is typical of this area of France, with low flows during summer and autumn, and a relatively short period of high discharge (70% of the annual flow) from December to March.

Nutrient loads were calculated using data from point sources and estimates based on agricultural statistics and land use. Export from each subcatchment and from the whole catchment was calculated using measured concentrations and flow rates.

One of the main features of phosphorus movements was the large amount retained in the Haute Vilaine reservoir (28% of input load retained). Overall, 15% of the total catchment phosphorus load was retained in the different reservoirs.

Within the study the quantity of total P input from the town's industrial and municipal sewage plants was calculated as 1.8 tonnes/year; the standard of wastewater treatment was not stated. This was considered to be insufficient to account for the load measured, so it was inferred that other local point sources may also contribute. Overall, total P export from the whole catchment was 103 tonnes-P over the year studied.

P loss from land varied from 1.15 to 1.3 kg total P/ha/year. Non point phosphorus sources (agriculture, presumably with a contribution from dispersed households not connected to a sewer) were estimated to be 90% of total P inputs. This is for an area with a low population density and

intensive agriculture. It is not typical of the Vilaine downstream of Rennes, whose conurbation has 272000 inhabitants (Statesman's Yearbook 2002) and whose input of P to the Vilaine river could be as big as the whole Haute Vilaine catchment.

3.3.7 Conclusions

In 1990, the implementation of the National Environment Plan and the detergent phosphate limitations imposed by the Environment Secretary were the first legislative measures introduced to reduce the amount of phosphorus in surface waters. Prior to this no policies or voluntary agreements were in place despite increasing public pressure to reduce eutrophication of water bodies.

The agreements between the Environment Secretary and detergent manufacturers, implemented in early 90s have resulted in a substantial reduction in the use of phosphates in detergents and encouraged the change to alternative, zeolite based detergents. While the limits set on phosphate concentrations were high, the manufacturers responded to public pressure by reducing the STPP content of their main products, and the market share of zeolite based detergents has risen 50% (table 3.2). These changes have coincided with the installation of phosphorus removal at some sewage treatment plants resulting from the implementation of the NEP (1990) and UWWTD (beginning in 1994 and still continuing). Subsequently, reduced phosphorus concentrations have been observed in major rivers in France since 1993.

In two catchments studied in detail (Redon river, Haute Vilaine) detergent phosphates were found to be a relatively minor component. Both these are rural areas where agriculture is important, and not typical of more urban catchments, where urban wastewater is the main source.

Although improvements were delayed due to the lack of legislative or voluntary control prior to the 1990s, it is evident that the combined approach of improved wastewater treatment and reduction in detergent phosphates since the early 1990s has reduced surface water phosphorus concentrations in France. However, further reductions are still needed.

References

Comité Environnement Détergents (1999) *A contribution to the evaluation of the environmental risks of different domestic laundry detergent components*. Available from: AISD (Association des Industries des Savons et des Détergents), 118 avenue Achille Peretti, 92200 Neuilly sur Seine, France.

EMPA (1999) *Life cycle inventories for the production of detergent ingredients*, EMPA report No. 244, EMPA, St. Gallen.

EUROSTAT (1995) *Europe's Environment – Statistical compendium for the Dobris assessment*, Eurostat Statistical Office of the European Communities, Luxembourg.

French Environment Ministry (FEM) (1986) *Studies of the Redon river and of phosphorus enrichment of Lake Geneva*, Report of FEM Phosphorus – Redon sub-working group, Paris.

French Ministry of Finance (FMF) (2000) 'Taxe generale sur les activites polluantes (TGAP)', *Bulletin officiel de douanes*, no. 6421, 4 April 2000.

IFEN (1999) *L'Environnement en France – 1999 Edition*. IFEN – Orleans.

IFEN (1999) L'eutrophisation des rivieres en France: ou est la pollution verte?

IFEN (2000) *Les indicateurs de performance environnementale de la France*. IFEN – Orleans. Available at: <http://www.ifen.fr/perf/perf2000/>

Key Note (1997) *Household soaps and detergents: 1997 Key Note plus market report 11th ed.* edited by Zoe Ratcliff, Hampton: Key Note Publications.

Köhler (2001) *Detergent phosphates and detergent ecotaxes: a policy assessment*. Centre Européen d'Etudes des Polyphosphates, CEFIC – Belgium.

Moreau, S. and Bertru, G. (1998) Seasonal and spatial trends of nitrogen and phosphorus loads to the upper catchment of the river Vilaine (Brittany): relationships with land use. *Hydrobiologia* 373/374.

OECD (1997) *Environmental Data Compendium 1997*, OECD, Paris.

3.4 Germany

3.4.1 Legislative Background

In Germany a similar pattern emerges to that of Switzerland in which phosphorus was defended initially on the ground that substitutes would be more expensive than alternative ways of reducing phosphorus discharges, such as better treatment of wastewater. In 1972 Henkel argued that the cost of introducing increased sewage treatment (2.50 DM/capita/year) would be far less to the consumer than changing the composition of detergents. A joint research programme between Henkel (who had held the patent for zeolite since 1973) and the German government resulted in production of zeolite being advocated on economic grounds, almost a decade later.

This research led to the regulation of phosphate content of detergents by the "Phosphate-Höchstmengenverordnung", which stipulates maximum concentrations, and which entered in to force on 1 January 1984. The maximum permitted concentration of phosphates in detergents was reduced by 50%. Following the regulation there was a decline in the consumption of STTP, from 185900 tonnes in 1984 to 13000 tonnes in 1990, and none in 1998 (table 2.5).

The significance of this legislation must also be viewed in a wider context. Other factors in explaining the reduction include voluntary agreements. The use of phosphate free-detergents was an industry led development, encouraged by public debate on the eutrophication of the aquatic environment. Since 1986 consumers have generally decided in favour of phosphate free products and since then there has been virtually no phosphates in detergents in Germany.

As well as negotiating with industry to reduce phosphorus concentrations, Germany has made a clear commitment to increasing the level of tertiary treatment. This commitment is illustrated in the Waste Water Charges Act (1981).

Under this scheme the Länder are able to collect levies on substances discharged in wastewater (including phosphorus) into water bodies. The amounts of such levies are based on the substance's degree of toxicity, and expressed in units of toxicity. The levies are imposed on all direct dischargers, i.e. they are imposed primarily on local authorities, as operators of public wastewater treatment facilities, and on large industrial facilities that have their own wastewater treatment installations.

The Act sets out in detail the load of phosphorus and other pollutants such as nitrogen, organic halogen compounds and mercury, that in each case, corresponds to one unit of toxicity. Three kg of phosphorus correspond to a unit of toxicity, compared to 1 kg Cu or 100 g Cd. Since the fee was introduced, the fee rate has been repeatedly increased. As of 1 January 1997, it is 70 DM per unit of toxicity.

The revenue generated by the fee goes to the Länder. It must be used for measures that support water quality of water bodies. In 1998, the total fee revenue amounted to 720 million DM, of which some 592 million DM was generated in the old Länder (western Germany). According to available data, the administrative costs amount to about 76 million DM.

International considerations have also played a part. Germany is one of the signatories to the Rhine Action programme, which required a 50% reduction in inputs of phosphorus and nitrogen to surface waters.

The Strategic Action Plan for the Danube sets short, medium and long term targets for implementing policies to reduce nutrient inputs from urban waste water treatment plants by 50% by 1995, industry and agriculture as a frame-work for national action plans. The Action Plan supports and complements the Danube Convention.

3.4.2 Changes in phosphorus discharges

Phosphorus discharges to rivers (Fließgewässer) was estimated by Hamm (1996) and quoted by Ecologic (2000). Hamm's estimates are shown in table 3.12 below.

Table 3-12 Estimates quantities of P discharged to German rivers (Hamm)

Source	1975	1985	1987/9	1995
Agriculture	27	31	35	24
Detergents	57	35	15	~0
Others	63	57	57	42
<i>Total</i>	<i>147</i>	<i>123</i>	<i>107</i>	<i>66</i>

Thousands of tonnes per year. 2.8 gP/person/day assumed for human waste excluding detergents.

Dr. Hamm, who is Chairman of the Main Committee of the German Scientists' Water Chemistry Group, also examined the effect on watercourses if detergent phosphates were to reappear in

what was West Germany. He estimated that given current (1996) wastewater treatment, the reintroduction of STPP based detergents would result in an increase in P discharged of 5-6000 tonnes/year.

These figures indicate that the change from STPP to Zeolite based detergents led to a reduction in total P discharged to German rivers of ~30000 tonnes/year in the late 1980s. Clearly the size of the reduction was related to the standard of sewage treatment at the time.

Behrendt et al (1999) estimate the quantities of phosphorus discharged to German rivers in 1985 and 1995. Their results are shown in table 3.13. They are considerably less than Hamm's estimates; Behrendt et al discuss possible reasons for the difference, which include using different values for the amount of phosphorus produced per person per day.

Table 3-13 Estimates quantities of P discharged to German rivers (Behrend et al)

Source	1985	1995
Municipal wastewater	56.9	11.4
Industrial point sources	7.1	1.3
Diffuse sources ¹	29.6	24.6
<i>Total</i>	<i>93.6</i>	<i>37.7</i>

Note 1: diffuse sources are ~70% agricultural.

1.9 gP/person/day assumed for human waste excluding detergents.

Both estimates agree on the major reduction in phosphorus discharged in treated wastewater between 1985 and 1995. Behrendt et al's results do not show the effect of the change to Zeolite based detergents explicitly; however their assumptions, and Hamm's results, are consistent with ca. 50% of the reduction being due to this cause, and 50% to improved wastewater treatment.

3.4.3 River water quality improvements

Rhine

The Department of Biological Oceanography of the Biologische Anstalt Helgoland has a number of data sets that are relevant for the evaluation of surface water eutrophication .

Phosphate levels increased from 0.5 mg/L in 1962 to 0.9 mg/L within 10 years but have decreased since then; the reported average concentration was 0.48 mg/L in 1985, falling to 0.15 mg/L in 1995 (Ecologic 2000 and fig 3.9). It appears that the abatement strategies for nutrients have been more successful for phosphorus than for nitrogen.

Figure 3.7 presents concentrations of orthophosphate and total phosphate in the Rhine river at Lobith, covering the period 1975-1998 (RIWA, 2000). The general trend for both variables is of decreasing concentration since throughout the duration of monitoring, with a gradual equilibrium being reached by the early 1990s. Lobith is located close to the Dutch/German national border and therefore surface water quality at this point is likely to reflect events in Germany.

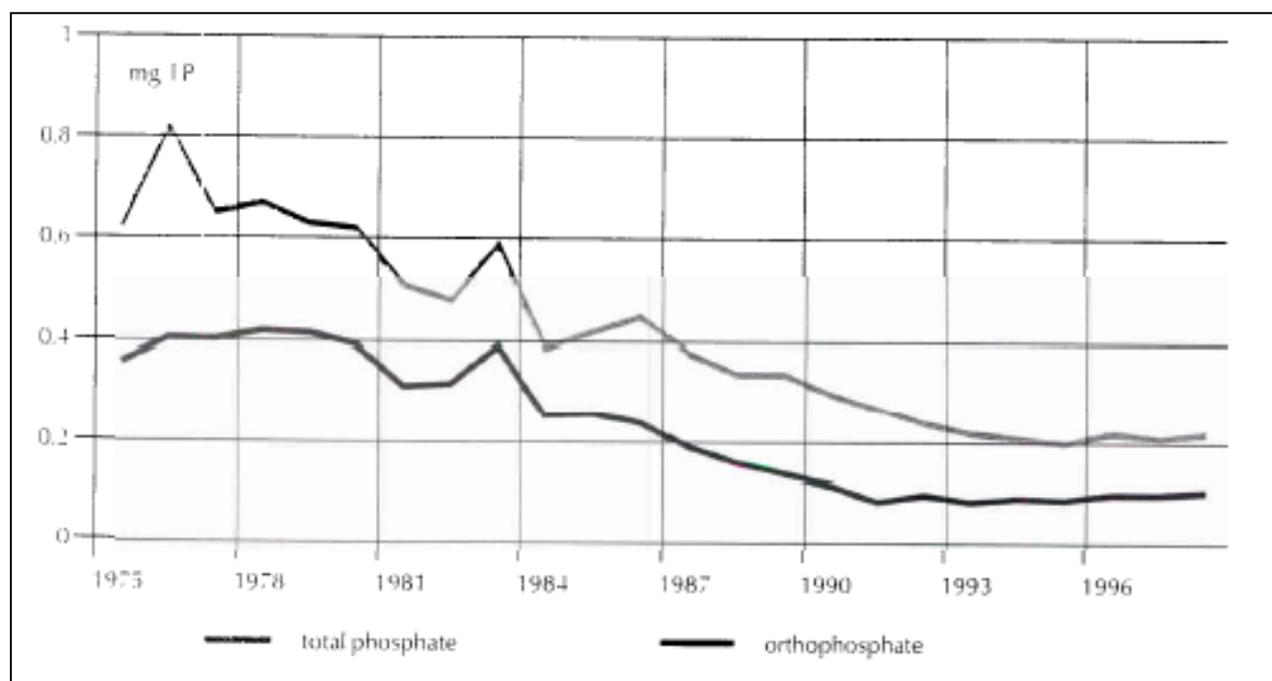


Figure 3.7 Concentrations of orthophosphate and total phosphate in Rhine water at Lobith, 1975-1998 (source: RIWA, 2000)

Improvements in the quality of the Rhine are reported by the EEA (undated), in terms of the dissolved oxygen concentration (at Bimmen), which rose from 5 mg/l in 1971 to 10 mg/l in 1995, and in the diversity of species, which was approaching its 1900-1920 level in 1995, after some decades with poor biodiversity. The EEA attribute this improvement in large measure to reduced BOD discharges. The effect of reduced phosphorus discharges is probably more apparent at the lower end of the Rhine, i.e. in Holland.

Lake Feldberger Haussee

Lake Haussee was originally considered to be only mildly eutrophic. However, large inputs of phosphorus and nitrogen from wastewater discharges during the sixties and seventies resulted in cultural eutrophication of the lake and subsequent blooms of green (chlorophytes) and blue-green (cyanophytes) algae were observed. In 1980 wastewater was diverted away from the drainage basin, leading to a 90% reduction in phosphorus inputs into the lake. It is well documented that lakes with a large nutrient pool often experience a long lag time in recovery following a reduction in external nutrient load (Koschel *et al* 1993). This was illustrated in Lake Haussee, which despite significant reductions in nutrient load in 1980, improvements in lake water quality were not observed until 1985 (Koschel *et al* 1993, Kreinitz *et al* 1996, Mehner *et al* 2001).

Due to this response lag-time, a biomanipulation experiment was initiated 1985 aimed at accelerating the recovery process. The first signs of restoration became evident between 1985-1989 with increases in crustacean grazers (daphnia) and decreases in nutrient concentrations (Kreinitz *et al* 1996). However, detailed analyses of the phytoplankton data revealed that non-heterocystis cyanobacteria dominated (90%) in summer months during these years, while

chlorophytes and bacillariophytes (diatoms) dominated during the previous years. Secchi depth (transparency) fluctuated throughout the study in response to changes in phytoplankton biomass. Between 1990 and 1992 the long term positive biomanipulation effects became apparent and phytoplankton biomass began to stabilise and increase in diversity (Kreintitz *et al* 1996).

Phosphorus concentrations in the lake decreased by 90% between 1985 and 1998 stabilising to around 100µg/L during the last few years of the experiment. These reductions occurred despite the fact that the external P load had not changed since the diversion of effluent in 1980. It was observed however, that the internal P loading decreased substantially after 1985. The key factors implicated in this reduction of P were:

- enhanced calcite precipitation and resultant phosphorus co-precipitation and sedimentation;
- decreased P mobilisation due to a decreased pH;
- increased N/P ratio; and,
- recolonisation of the littoral zone with macrophytes.

The study demonstrated that the various reactions between the chemical fluxes and phytoplankton assemblages resulted of interactions between both the 'top down' (food web manipulation) and 'bottom up' (nutrient loading) processes. Therefore it was concluded that the long term recovery of the lake was a result of a combination of reductions in the external nutrient load and appropriate long term manipulation of food web community structure, through reductions in planktivorous fish and subsequent increases in herbivorous zooplankton (Mehner *et al* 2001).

Conclusions

The research by Henkel and the German Government led to the limiting of phosphorus concentrations in 1980 with the aim of reducing by 50% by 1984. As in the USA, NTA products were released in the early 1980s but later withdrawn following safety concerns, so that by 1983 the first phosphorus free detergents using zeolite were on the market. Four years after their release, zeolite based detergents accounted for 50% of the market.

This reduction in detergent phosphorus, along with improved wastewater treatment resulted in a substantial reduction in phosphorus reaching watercourses from urban wastewater between the period 1974-1989, and this figure was further reduced as a result of the UWWTD and continuing use of P-free detergents. These reductions have subsequently been effective in reducing total phosphorus concentrations in the Rhine from between 0.6-0.8 mg/L in the 1970's to around 0.15 mg/L in 1995. Lake Haussee has also exhibited signs of restoration due to a combination of a reduction in the external nutrient load and biomanipulation experiments.

3.4.4 References

Koschel, R.; Kasprzak, P.; Krienitz, L. & Ronneberger, D. (1993): Long-term effects by reducing of nutrient loading and following food-web manipulation in a stratified Baltic hardwater lake (Lake Haussee, Germany). *Verh. Internat. Verein. Limnol.* 25, 647-651.

Krienitz, L. Kasprzak, P. & Koschel, R. (1996): Long term study on the influence of eutrophication, restoration and biomanipulation on the structure and development of phytoplankton communities in Feldberger Haussee (Baltic Lake District, Germany). *Hydrobiologia* 330, 89-110

Mehner, T., Kasprzak, P, Wysujack, K., Laude, U. & Koschel, R. (2001): Restoration of a stratified lake (Feldberger Haussee, Germany) by a combination of nutrient load reduction and long-term biomanipulation. *Internat. Rev. Hydrobiol.*, 86, 253-265

3.5 Hungary

3.5.1 Hungary, the Danube and the Black Sea

The Black Sea is virtually isolated, being connected with the Mediterranean by the narrow Bosphorus channel. Some parts are deep, but the North Western shelf is relatively shallow (<200m deep).

At least 162 million people live in the Black Sea basin (SENATOR 1996), of whom 50% live in the Danube Basin. The Danube is the biggest river entering the Black Sea, and contributes 75% of the flow to its North Western shelf. The quality of the Danube is therefore a critical influence on the quality of the NW shelf, which is currently eutrophic.

Hungary is one of 13 nations with at least part of their area in the Danube catchment. It comprises 11% of the total catchment area of 818000 km², and 12% of the total population of 85 million, and is the second largest to Romania on both counts.

Eutrophication problems in Hungary itself are relatively local, for example at Lake Balaton (SENATOR 1996), and the Hungarian public may not be directly aware of the impact of their actions on the Black Sea.

3.5.2 Legislative Background

Hungary has in place a system of fines for non-compliance of treated sewage effluent with standards. Fines are levied in according to a formula that takes into account the degree to which effluent limits are exceeded. The area limit values for Phosphorus are 1.8 and 2 mg/L. Due to the inability or unwillingness of companies to pay the fines, combined with the relatively high cost of capital investment required to meet the standards, the system has proved difficult to implement.

Limits on the phosphate content in detergents were introduced by the "Standard for Pulverous Synthetic Detergents" in 1986. The revised standard for the determination of total phosphate content in detergents was released in 1987.

The Standard classified detergents as follows:

		P ₂ O ₂ content
A	Environmentally friendly	<7%
B	Within safe limit	7 > 15%
C	Max allowable	Max. 20%
D	Prohibited	>20%

There is also a voluntary agreement between Government and manufacturers to reduce detergent use and improve public awareness.

Despite these restrictions the Environmental Programme for the Danube River Basin identified the high levels of nutrients as one of the main problems affecting the health of the Danube in its outline of the strategic Action Plan for the Danube River Basin. As part of the integrated policy approach of this plan one of the stated goals is the banning of phosphates in detergents by 2005 along with measures aimed at reducing the influence of agricultural run-off.

Hungary is also a signatory of the Strategic Action Plan for the rehabilitation and Protection of the Black Sea (1996) the goals of which include:

- reduction of nutrient loads in rivers until Black Sea water quality objectives are met
- reduction of pollution from point sources by 2006: first progress report required by 2001
- each Black Sea state to develop National Strategic Plan for point source reduction
- significant reduction of inputs of insufficiently treated sewage from large urban areas by 2006

3.5.3 Detergent use trends

Phosphate-free detergents are used in relatively small quantities in much of the CEE, in the Czech Republic (35% in 1998, table 2.5), Hungary (30% in 1998), but this proportion appears to be rising. As in western Europe zeolite A is the most common alternative.

3.5.4 Wastewater collection and treatment

Currently only 50% of the population is connected to a sewer system. Planned investments are therefore mainly in new sewer systems, and secondary biological wastewater treatment. Currently Hungary has identified as sensitive waters some lakes including lake Balaton, but not the Danube or its tributaries. Therefore improvement plans are aimed at sewer system construction and secondary sewage treatment, but not generally nutrient removal.

Table 3-14 Wastewater collection and treatment levels – Hungary, 2001

	Total		With collecting systems		With wastewater treatment	
	PE 000s	Number	PE 000s	Number	Est. PE 000s	Number
<2		1.91		0.03		0.03
2 to 10	334	1.59	187	0.25	173	0.24
10 to 15	57	0.70	53	0.19	50	0.20
15 to 150	111	4.48	109	2.68	97	2.34
>150	10	6.01	10	5.50	8	1.85
Total		14.70		8.65		4.66

Source of data: Hungarian National Programme for the Implementation of the Council Directive 91/271 EEC Concerning Urban Waste Water Collection and Treatment, Hungarian Ministry of Transport and Water Management, February 2001. There is minimal nutrient removal (table C.1).

3.5.5 Surface water quality

Of particular concern has been the increase in the nutrient load to the Black Sea in the past 25 years, which is well documented. Since 1969 annual mean concentrations of phosphorus on the North West shelf have risen from less than 0.05 mg/l to over 0.15 mg/l in 1985, and are still high. This area is eutrophic.

Although the concentration of total P in the Danube in Hungary is high (>0.2 mg/L in 1992), eutrophication is not apparent there.

Lake Balaton is Hungary's major inland lake. Eutrophication has occurred and continues to occur despite a reduction of ~50% in the [P] load entering the lake. This was attributed to recycling of P in the sediments (Clement et al. 1998).

3.5.6 Phosphorus discharges

Detailed data regarding the historic trend of phosphorus fluxes in Hungary is not readily available, however Haskoning (1994) reports approximately 8,200 t/P year to surface waters in 1991. Most of this is from point sources (table 3.15), although for the Danube as a whole, agriculture is the largest source.

Data for the International Commission for the Protection of the Danube River indicates that in 1997 industrial and municipal sources in Hungary were responsible for 3850 t/ phosphorus entering the Danube.

Table 3-15 Sources of Phosphorus in the Danube Basin

	Whole basin	Hungary	
	%	000 t/yr	%
Population	27.2	5.5	67
Agriculture	53.2	1.1	13
Industry and atmosphere	19.6	1.6	20
Total		8.2	

Sources: Haskoning 1994, EPDRB

The EPDRB (Environmental Programme for the Danube River Basin) have produced estimates of future phosphorus discharges to surface waters based on various scenarios of treatment plant development and phosphorus removal from detergents Hungary. Their estimates for the population and industry were similar to Haskoning's, but that for agriculture was higher at 7000 t/yr. Estimates of phosphorus inputs from agricultural activities are uncertain, but may have decreased since 1990 as the quantity of P fertiliser applied to land has fallen (Haskoning 1994).

SENATOR considered three main scenarios:

Scenario A Autonomous development

Scenario B 50% reduction of P in detergents

Scenario C 100% reduction from detergents

Within these three main scenarios the various sub-scenarios were considered:

1. Only those treatment plants in operation that are in place in 1994 with no additional removal of phosphorus from detergents
2. As 1, and removal of Nutrients as in line with EU Directives
3. Treatment according the Hungarian standards

Table 3-16 Estimated quantities of total P from population discharged to Hungarian surface waters, 2010

Detergent options	Sewerage & sewage treatment options		
	1. No additional sewage treatment or P removal	2. Nutrient removal in line with EU directive	3. Hungarian standard ¹
A. 100% STPP	7.27/2.47	6.04/1.24	4.80/0.00
B. 50% STPP, 50% Zeolite	5.23/1.83	4.31/0.91	3.40/0.00
C. 100% Zeolite	3.86/1.35	3.19/0.68	2.51/0.00

Note 1. This provides for nutrient removal at all sewage treatment works, and is probably unrealistic.

Note 2. Quantities are in kt/year. The first figure is the total, and the second the contribution from detergents.

Note 3. The base line values for 1994 were 4.82/1.65.

As **table 3.16** illustrates there are a number of combinations of policies which would achieve the same level of Phosphorus discharged to surface waters. For example, a complete change to

Zeolite as a detergent builder (1.C) would achieve the same as nutrient removal at all treatment works with the current mix of detergent builders (70% STPP, 30% Zeolite, intermediate between 3.A and 3.B). The cost of the former to the Hungarian economy would be small; the latter scenario would cost an estimated 4 to 6 billion euros, or several hundred euros per person (SENATOR 1996 p.139). Furthermore Zeissner et al (1998) point out that the unit costs of sewerage and sewage treatment are two to four times as high for small communities (2000 to 10000 people) as for large communities (>100000 people). Small communities in Accession States such as Hungary will find it difficult to afford these investments.

Specific targets for reductions in P discharges to the Danube appear not to have been set in the Danube Convention and subsequent documents. However the current level of total P in the NW Black Sea (>~0.1 mg/L) suggests that reductions of at least 50% and probably more will be needed. Neither a change to Zeolite as detergent builder, nor improved sewage treatment, will be sufficient by itself. Both are necessary.

Conclusion

Hungary is an example of the particular problems faced by the accession countries, in particular the low proportion of the population connected to treatment works and the compounding factor of low levels of treatment for the proportion that is collected. It is likely to take more than a decade to achieve the UWWTD standards.

In this context, reducing the phosphorus content of detergents is a cost effective means of reducing phosphorus inputs to surface water.

To control eutrophication in the NW Black Sea, both actions are necessary.

3.6 Italy

3.6.1 Legislation and official measures

Two continuing issues in Italian legislation have been detergents and eutrophication. Water quality problems have been obvious to the public since the 1970s, and have affected commercial interests, in particular tourism and fishing (including shell fisheries).

The legislation on detergents has covered the use of NTA, surfactants, and the biodegradability of their constituents. The two laws shown here are those that apply in particular to the phosphorus content.

Decree of 30.12.81, nr.801, brought into law 29.05.82	The phosphorus content of detergents was limited to 5% from 24 months after entry of	
---	--	--

nr.308	the law into force. The % of phosphorus had to be shown on packets and containers.	
13.09.88, nr.413	From 1 January 1989, detergents (except for those for dishwashers) may not contain more than 1% phosphorus expressed as P	This was introduced following advice from an expert committee, chaired by the Ministry of Health and with representatives of national and regional government and the relevant industrial sectors

Eutrophication has been a continuing issue, with laws or decrees containing 'urgent measures' being issued at a national level in 1986 (24.01.86, nr.7) and 1989 (04.08.89, nr.283). The 1988 law limiting the P content of detergents to 1% was one of the measures aimed at controlling eutrophication.

In the 1990s the Po River Basin Authority actively considered pollution sources and ways of improving river quality; see for example the two measures approved below by the Autorita di Bacino del Fiume Po (Po river basin authority).

15.04.96, nr.12/96	Approval of a directive for containing the pollution from animal husbandry in the Po basin
14.10.98, nr.24/98	Approval of urgent measures to combat eutrophication in inland waters and the Adriatic Sea

3.6.2 Detergent constituents

Between 1985 and 1989, there was a change from predominantly STPP based detergents to Zeolite A based laundry detergents (table 3.2), corresponding to the law of 13.09.88 coming into force in January 1989.

3.6.3 Northern Italian lakes

The region of Lombardy recently carried out a review of the environment generally, with one chapter on water quality (Regione Lombardia, undated). In it, the quality of several lakes is summarised but no history is given. In its review, the Regione Lombardia states the following:

- 92% of the population is connected to a sewer system, compared to 78% in 1985. Most of the sewers are combined, i.e. they carry rain water as well as foul sewage. Overflows are a

source of pollution. the plan provides for 99% of the population to be connected. The majority of the sewers will remain combined.

- The region plans to rationalise sewage treatment, creating larger treatment centres and reducing the number of treatment works serving small populations.

Details of two particular lakes are given below.

Lago d'Iseo (Garibaldi et al 1998)

Lago d'Iseo is situated north of Bergamo. It has an average width of 2.5 km and a perimeter of 60 km. Its drainage basin is mountainous, with an average altitude of over 1000 m; it has 168000 residents, with a peak holiday population of 246000.

Garibaldi et al compared estimates of phosphorus inputs to the lake, and concentrations in the lake, with measured values.

The inputs of phosphorus are from:

- Two large sewage treatment works at the northern and southern ends:

	Northern	Southern
Current load (pe)	40000	22500
Current processes	Secondary biological treatment, P removal with lime	Not stated
Planned capacity	80000	90000
Discharge point	Lago d'Iseo	River Oglio downstream of the lake

- Other small sewage treatment works along the lake side. These do not remove phosphorus.
- Isolated dwellings.
- Industrial sources. There are none that discharge large quantities of phosphorus; 10% of the domestic load was assumed.
- Agricultural run off and animal wastes. ISTAT data were used to estimate loads. There is no intensive animal husbandry.

Three cases were considered:

- Past. Garibaldi et al do not define this, but presumably detergents were assumed to contain STPP and sewage treatment was to a lower standard.
- Present,
- Future, assuming 9 more communes to be connected to the two main treatment works.

Good agreement between measured and estimated inputs was obtained. However the theoretical P concentration was much lower than that measured. The difference was attributed to phosphorus being released from the sediments under anoxic conditions; recently two layers have

persisted in the lake, a surface layer saturated with oxygen and a deep anaerobic layer in contact with the sediment.

Garibaldi et al conclude that further reductions in P inputs are necessary, by at least 20 tonnes/year beyond the 'future' scenario, and suggest that sewage should be collected from the small sewage treatment plants and treated at the southern end of the lake where there is spare treatment capacity.

	Total P entering the lake (tonnes/year P)	Point & diffuse sources	Average P concentration (ug/l)
Past	Estimated 200		Estimated ¹ 31-36
Present (1996/7)	Estimated 90 Measured 80	45% point, 55% diffuse	Estimated ¹ 17-18 Measured 55
Future	Estimated 80		Estimated ¹ 14-15

Note 1. Estimated using the 1982 OECD model.

Lago Endine (Garibaldi et al 1997)

Lago Endine is situated to the west of the Lago d'Iseo, and north east of Bergamo.

Its area is 2.34 km², length 6.08 km and average depth 5.1 m. The drainage basin is described as 'pre-alpine', with a mean altitude of 685 m and a maximum of 1381 m. The total population is 6000, with 2700 summer visitors. Industrial and agricultural discharges have not been, and are not now, important sources of nutrients.

A summary of the lake's history is given in [table 3.17](#). There are two main factors causing the reduction in P load entering the lake: construction of the collector sewer, and the change in detergent builder between 1986 and 1992.

Garibaldi et al concluded (in 1997):

'...the improved conditions of the last two years are not enough to sustain the complete recovery of the lake; rather, the present condition of the lake is a delicate passage from eutrophy to mesotrophy. A key variable at this stage is the reduction of the internal load, as the amount of phosphorus stored in the sediments during the last few years is substantial.'

Table 3-17 Lake Endine history

Year	Estimated total P input (as P)	Total P concentration (ug/l)	State of the lake	Action
1973	10.2 t/year per year. Also high internal load from sediments, released during anoxic periods.		Highly trophic.	Regular twice yearly sampling
1978				Detailed study by Bonomo et al. Recommendation for treatment of all point sources of sewage, by means of a 'ring' collector sewer and a treatment plant at the lake outflow.
1981	10.2 t/year	Average >70?		
1984-5		Peak 150-200		
1986	6.75 t/year	Average 40		
1989				Limit on P in laundry detergents of 1% becomes effective
1992	2.14 t/year	Average 30 Peak 120		
1996	1.80 t/year. Lower release of P from sediments.	Average 20 Peak 50	Reduced anoxia. Transition to mesotrophic state begins.	Ring sewer 80% complete, with the main town, Endine, connected. Sewage is taken to Trescore Balneario, 10 km downstream.
2000? Planned		Target average 10		Completion of the ring sewer Construction of sewage treatment at Trescore Balneario

3.6.4 Northern Adriatic (Regione Emilia Romagna, 2002, Nasci et al, CNR)

The Adriatic sea is surrounded by the Italian peninsula to the West and by the Balkan peninsula to the East and it is linked to Mediterranean sea through the Otranto strait.

It has a length of 800 km, an average width of 180 km, an area of 139000 km².

The sea is divided into three different basins with decreasing depth from south (70 m) to north (30m). The Northern basin is most vulnerable to human impact, mainly because of the density of population and to the high degree of industrialization of its Western coastal area.

In the Northern Western area the fluvial deposits of the Po are the most important with a medium flow of 1500 m³ s⁻¹. This freshwater flow generates conditions of great fertility and richness of fish resources in the high Adriatic Italian coast. The flow of water to the sea can treble after high rainfall and this implies the introduction of high masses of nutrients into the sea.

The quality of the Adriatic north of Ravenna is strongly influenced by inputs from the river Po. The estimated load of phosphorus entering the Adriatic is 10750 t/year total P. Most of this (56%) was estimated to be from domestic wastewater (Po river basin authority, 1998).

From 1975 onwards, following intense growth of algae and phytoplankton, anoxic zones occurred resulting in the death of benthic organisms and their being washed up on beaches. The tourism and fishing industries suffered as a result.

Improvements in sewage treatment in the Po catchment have been made in the 1980s and 1990s (Regione Lombardia, undated), although not consistently throughout the basin. For example, after 15 years of planning, the city of Milan (population 3 million) still has no sewage treatment. After the mayor of Milan admitted the impossibility of completing sewage treatment by the agreed date of 31.12.2000, the national government set up a special commission to oversee the project (Ord Prot Civile 3041/2000, Gazzetta Ufficiale 52 of 3.12.2000). Most large cities have secondary sewage treatment, but several, e.g. Padova and Venezia, do not have nutrient removal (European Commission 2001).

From 1988 onwards the summer micro-algal blooms reduced in intensity, extent and duration. This has been measured by the trophic index which takes account of four parameters commonly used in the study of eutrophication: chlorophyll a, dissolved oxygen, dissolved inorganic nitrogen and total phosphorus. The reduction in phosphorus concentration [from 0.28 to 0.18 mg/l or 36%] is one of the factors that has brought about this improvement. Further reduction is needed, as damage to fishing and tourism still occurs (Po river basin authority, 1998), and the Po authority has set a target of 0.12 mg P/l at the mouth of the Po for 2008, and 0.10 for 2016.

	Total P	Trophic index ²		
	Po river ¹	P. Garibaldi	Cesenatico	Cattolica
1983-87	0.2 to 0.36, average 0.28	6 to 6.5	5.5 to 6.5	5 to 5.5
1988-95	0.06 to 0.23, average 0.18	5.6 to 6	5.2 to 5.8	4.7 to 5.2 (from 1990)

Source Caiaffa 1999

Note 1. Po river station (It8 Ponte lago scuro), average total P concentrations (mg/l)

Note 2. At 3 points along the Adriatic coast. P.Garibaldi is near the mouth of the Po, Cattolica is furthest away to the south.

Note 3. A Trophic index of 5 to 6 corresponds to a bad trophic state. Less than 5 is classed as good.

A different phenomenon is that of 'mucillagini', arising from growth of diatoms over many square kilometres of sea. These occur elsewhere in the Mediterranean, but are of particular concern in the Adriatic because of its shallowness. Recent peaks occurred in 1988, 1989, 1991 and 1997. The causes have not yet been confirmed, and are the subject of continuing study.

3.6.5 Conclusions

In Italy, public concern over water quality, and in particular eutrophication, began in the 1970s. Commercial interests were affected, in particular tourism and fisheries. These pressures led to the limiting of P content of laundry detergents to 1% from 1/1/1989; since that date laundry detergents have contained Zeolite A as the main builder. As a result of this, and patchy improvement in sewage treatment, there has been a reduction on the P concentration of the northern Adriatic by 30-40%, and an improvement in the eutrophic index.

Further improvement is still needed in the Adriatic, and to help achieve this the Po basin authority has set a target concentration of 0.10 mg P/l at the mouth of the Po, representing a reduction of 65% compared to the 1970s.

The histories of the two lakes – Iseo and Endine – illustrate the need to reduce P inputs by 60% or more compared to the 1970s, and the role of sediments in maintaining high phosphorus concentrations after P inputs have been reduced.

3.6.6 References

Bonomo *et al* (1978). *Alternativa tra collettore circumlacuale e impianti autonomi per il risanamento del Lago d'Endine*, Ingegneria Ambientale 7 705-716

Caiaffa (1999). *European Marine Information System*, paper presented at the Inrer-Regional Forum of European Conventions, September 1999

Garibaldi L *et al* (1997). *The improving trophic conditions of Lake Endine*, Mem Ist ital Idrobiol 56 23-36

Garibaldi *et al* (1998). *Apporti di fosforo al Sebino*. Evoluzione idrochimica a trofica del Lago Iseo, Documenta Ist ital Idrobiol 53 191-212

Po river basin authority, (1998). *Piano delle direttive ed interventi urgenti per la lotta all'eutrofizzazione delle acque interne e dell'Adriatico*

Po river basin authority, (2001). *Progetto di Piano stralcio per il controllo dell'Eutrofizzazione*

Regione Emilia Romagna (2002). *problemi ambientali dell'Emilia Romagna (on web site)*

Regione Friuli Venezia Giulia, (2000). *Riserva Naturale Marina di Miramare, Rapporto Modulo 3 (with WWF Italia)*

Regione Lombardia (undated). Regione Lombardia, DG Tutela Ambiente, Capitolo 8, Il Ciclo delle Acque

3.7 Netherlands

Key dates and impacts

- Pre 1987: Eutrophication highlighted as problem, but no statutory emission limits set for phosphorus;
- 1987: Rhine Action Plan recommends a 50% reduction of phosphates (1985 emission levels) by 1995. This translates to an WWTP efficiency improvement of 75%. Action was initiated by water boards immediately;
- 1987: Report from Dutch Ministry of Housing, Spatial Planning and the Environment (VROM) published, introducing a new instrument, the covenant, in which all concerned parties decided, on a voluntary basis, to reduce the amount of phosphates in washing powder.
- 1990: Fosfaatbesluit (Phosphates Decree) published. Existing phosphate reduction recommendations become statutory regulations for emissions from communal (public) waste water treatment plants;
- 1990: 'Voluntary Plan of Action – Laundry and Cleaning Products for Households' agreed.
- 1992: Netherlands national eco-labelling scheme established. Products labelled include detergents, which have to meet certain phosphate-content criteria.
- 1995: Reduction targets for phosphates met.
- 1996: UWWT Directive transposed into Dutch national law, three years later than the 1993 deadline. Minimal change to regulations already in place.

3.7.1 Legislative Background

When eutrophication first became a political issue, it was at first limited to phosphate pollution. In policy papers from 1975 onwards, water boards were encouraged to aim towards reducing phosphate emission loads from WWTPs, but no statutory emission limit was set. This non-binding target was consequently not reached. The lack of implemented measures to remove phosphates from urban waste water largely stemmed from the belief in the water sector that measures would not be cost effective (Kelder, 2000) as long as imported nutrient loads remained high (e.g. downstream from Germany) and phosphates were originating largely from detergents.

From 1970 to 1987 no statutory emission limits were set. This was predominantly due to phosphate treatment at WWTPs being inadequate, even when combined with the reduction of phosphates used in washing powder. Although eutrophication processes had been observed within the Netherlands throughout the 1970s and early 1980s – for example chlorophyll A concentrations in the IJsselmeer were normally above 50 ug/l and reached peaks every year above 200 ug/l - the Dutch Government did not act until 1988, after international agreements were achieved.

International agreements

The Netherlands expressed interest in international agreements on phosphate reduction targets predominantly due to its geographical situation. In the delta of three major European rivers (Rhine, Meuse and Scheldt), more than 50% of the country lies below sea level, and 15% of the surface area consists of water: rivers, lakes, pools, canals, brooks and marshes. In 1996 the phosphate load imported from adjacent countries was 1.5 times the inland load (although a large component of the imported loads are transported directly to the sea) (Kelder, 2000). It is therefore of great importance to the Netherlands that international water management agreements occur at the European level.

The Rhine Action Plan (IKSR, Rhine Action Programme, Strasbourg, 1987) was aimed at reduction of phosphates in the River Rhine, after international agreement was achieved. The recommended 50% reduction of phosphates translated to an WWTP-plant efficiency improvement of 75%. In the light of the report 'Scenarios for Dephosphatation at WWTP-plants' (CUWVO, 1988), the water boards recommended that the requirements (emission limits and reduction targets) be translated into statutory regulation. The result was the *Fosfaatbesluit* 1990 (Phosphates Decree 1990), regulating emissions from communal (public) waste water treatment plants. Although emission limit values (ELVs) apply to the whole country, targets have to be reached within water board regions, giving the water boards the opportunity to keep older, less efficient WWTPs if newer WWTPs within the same region make up for the shortfall.

UWWT Directive

Statistics of waste-water treatment in Europe reveal that 69% of the population of the Netherlands were connected to a WWTP with secondary treatment (P-removal 25-30%) in 1997 (RIONED 1998) and 28% of households to be connected to a WWTP with tertiary treatment (P-removal >80%) (OECD, 1997 and Eurostat, 1995). Now (2002) a much higher proportion of wastewater is treated to remove phosphorus (table C.4 shows an estimate of over 90%).

The transposition of the Urban Waste Water Treatment (UWWT) Directive into Dutch law was completed on 1 March 1996, nearly three years after the deadline of July 1993. However Dutch legislation already implemented the directive to a substantial extent.

When the UWWT was introduced, the existing *Fosfaatbesluit* was completely integrated into the new *Lozingenbesluit Wvo stedelijk afvalwater* of 1996 (Decree on Discharging Urban Wastewater), although some of the existing regulations were stricter than the Directive and were allowed to remain. Among other things, this Decree prescribes requirements for obtaining a permit to discharge water via a wastewater treatment plant and includes guidelines on total-P discharges. An important requirement of the UWWT Directive is the designation of Sensitive Areas (Article 5), in response to which the Dutch Government announced that the whole area of the Netherlands would be considered sensitive, due to the geographical situation of the country. The effect of the announcement is such that all discharges of agglomerations greater than 10,000 pe should be treated for phosphates and nitrogen, although these requirements were already present in the previous decrees.

Reduction in phosphate content of detergents

In 1987 a phosphate report from VROM in 1987 introduced a new instrument, the covenant, in which the parties involved (including detergent manufacturers) decided, on a voluntary basis, to reduce the amount of phosphates in washing powder. This agreement resulted in almost all laundry detergents becoming phosphate free (95% by 1989, see table 3.2), thereby reducing the amount of phosphate in domestic wastewater by probably 40%.

The 'Voluntary Plan of Action – Laundry and Cleaning Products for Households' between the Dutch Ministry of Housing, Spatial Planning and the Environment (VROM) and the Dutch Soap Association (NVZ) was agreed in 1990. This plan facilitated discussions and co-operation on controversial issues such as the difference in views regarding risk assessment, risk management, and problems associated with chemical groups including phosphates.

Currently effectively all domestic laundry detergents used in the Netherlands have Zeolite A as a builder.

3.7.2 Surface water quality improvements

Significant reductions in the inputs of phosphate to surface water have been achieved since the introduction of measures to address the problem (see table 3.18). The Rhine Action Programme calls for a 50% reduction between 1985 and 1995, which was achieved. Targets for further reductions have been set.

Table 3-18 Phosphate (total-P) pollution of surface water in the Netherlands, 1985 – 1995, in 1000 ton/year (source: RIZA)

	1985	1995	% reduction
Industry (direct)	12.8	3.34	74
WWTP (effluent)	10.8	3.54	67
Diffuse sources	9.5	7.82	18
Total	33.1	14.7	56

Lake IJsselmeer

Lake IJsselmeer has a surface area of 1130 km² and an average depth of 4.3 m. About 80% of its water comes from the Rhine. The quality of water in the IJsselmeer has steadily improved since 1975, with total phosphorus concentration decreasing to approximately 40% of 1975 levels by 1995 (see figure 3.8). Rapid improvement was observed between 1985 and 1990, suggesting that the move to zeolite based detergents was a major factor. Chlorophyll *a* concentrations decreased by 10% between 1985 and 1995, much less than the decrease in total P. This may be due to the release of sediment phosphorus, or to a reduction in toxic substances that were inhibiting algal growth.

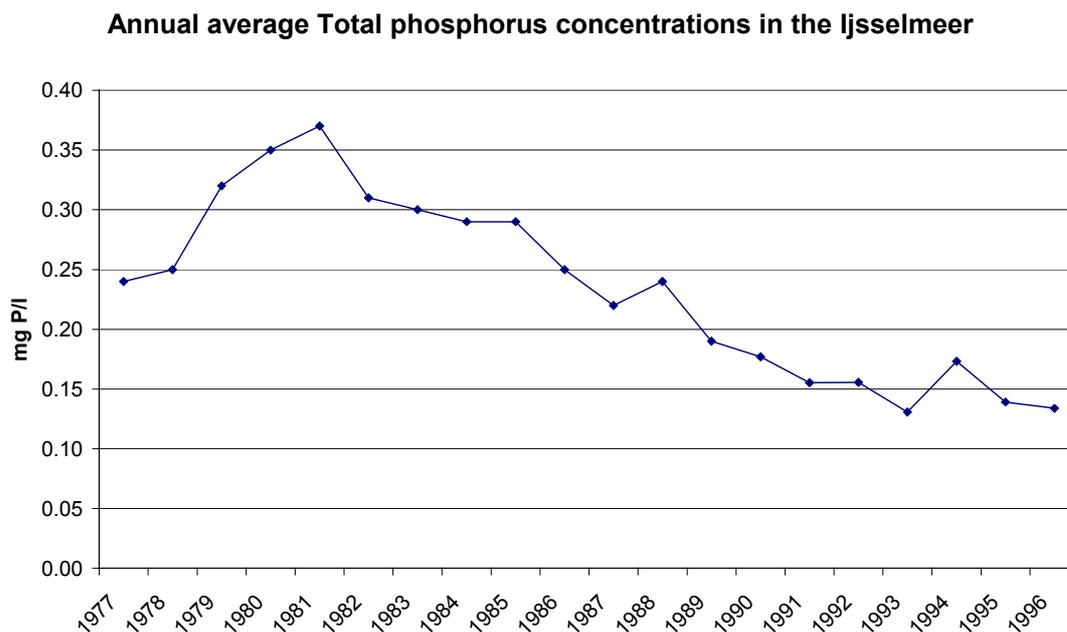


Figure 3.8 Total P trend in the IJsselmeer (Source: ETC/IW)

River Meuse

Figure 3.9, of total phosphorus concentrations in the River Meuse, displays a similarly decreasing trend in total P.



Figure 3.9 Total phosphorus concentrations monitored in the River Meuse at Keizersveer, 1977-1995 (Source: Data as reported to ETC-Inland Waters)

3.8 Conclusions

A voluntary agreement to reduce the levels of phosphorus in detergents was reached in 1987, and resulted in >95% replacement of STPP with Zeolite A. Additionally, substantial improvements in wastewater treatment were undertaken with the introduction of the Rhine Action Plan. These measures resulted in a reduction in phosphorus loads from wastewater effluent of over 60%.

Total Phosphorus concentrations in Lake IJsselmeer and the River Meuse decreased by 50% between 1985 and 1995; the relative importance of diffuse agricultural sources increased at the same time. However chlorophyll A concentrations in Lake IJsselmeer decreased by only 10% during the same time period.

References

AISE (1996) Environmental Risk Assessment of Detergent Chemicals – Proceedings of the AISE/CESIO Limelette III Workshop on 28-29 November 1995, AISE Brussels.

CUWVO (1988) *Coördinatiecommissie uitvoering wet verontreiniging oppervlaktewateren werkgroep actieplan defosfateren, Scenario's voor fosfaatverwijdering op rwzi's*, CUWVO.

Eurostat (1995) Europe's Environment – Statistical compendium for the Dobris assessment, Eurostat Statistical office of the European Communities, Luxembourg.

Kelder, E. (2000) *National Case Study on Policy Networks and Implementation of the Urban Waste Water Treatment Directive 91/271/EEC in the Netherlands – Final Report for TEP project*, Maastricht Economic Research Institute on Innovation and Technology (MERIT), Maastricht University, Netherlands.

OECD (1997) *Environmental Data Compendium 1997*, OECD, Paris.

RIONED (1998) *Het Riool in Cijfers 1998/99*, Stichting RIONED.

RIWA (2000) *Annual Report 1998, Part C: The Rhine and the Meuse*, International Association of River Waterworks, Amsterdam.

UBA (2000) *European Eco-Label: Revision of Eco-label criteria on laundry detergents*, Project No. 97/609/3040/DEB/E4, Parts I and II, Umweltbundesamt Berlin.

3.9 Switzerland

3.9.1 Legislative Background

The detrimental effect phosphates were having on Swiss waters was first recognised in the 1950s, and in 1955 the Water Protection Law initiated the construction of wastewater treatment plants and a more effective sewage system. At this time reduction of phosphorus at source was seen as only one of the requirements to improve Swiss waters.

By the early 1960's the legislative body had begun to impose restrictions on components of detergents. In 1961 an article of authorisation was added to the Water Protection Law issuing regulations on substances entering water. These regulations were later revised in 1971 and 1991.

Despite the recognition of the contribution of phosphates in detergents, no restrictions on phosphates were issued in the amendments of 1971. This was partly due to concerns regarding substitutes to phosphates, namely NTA and Zeolites. These concerns were expressed officially through the Federal Councils response to parliamentary questions in 1968. The introduction of further restrictions on the use of phosphates was also delayed by the unsuccessful introduction of phosphate free detergents in the early 1970s, which failed to deliver satisfactory cleaning standards.

In conjunction with the growing recognition of the need to limit phosphates at source was the recognition of the need for better treatment of wastewater. This led in 1967 to the Dept. of Interior issuing recommendations that a third level of treatment be installed specifically for the removal of phosphates in domestic wastewater within the catchment area of all lakes.

The increasing pressure on phosphate use led to a voluntary industry agreement restricting the amount of phosphates in detergents. Despite this agreement declining concentrations of phosphate were not seen until the late 1970s.

From 1981 onwards, further action was taken to restrict phosphorus in detergents, culminating in the Federal ban in 1985.

Table 3-19 Summary of Development of Legislation in Switzerland

1961	Federal Council decision to asses measures to counter the effects of synthetic detergents
1962	Creation of Commission on Detergents
1964-1968	Parliamentary pressure continues-Parliamentary questions enquiring about the phosphorus legislation in preparation.
1964-1968	No legislation is planned due to lack of viable alternatives to phosphorus
1967	Federal Dept. of Interior Introduce phosphorus removal at treatment plants across all Cantons
1969	Commission on detergents reports-no phosphorus ban
1971	Supplement to Water Protection Law regulations on products entering water
1972	Regulations issued on compounds in detergents but no restrictions on phosphorus content
1974	Voluntary agreement with industry limiting amount of phosphate
1977	Ordinance of detergents; maximum amounts of phosphates
1981/1983	Step wise tightening of phosphate concentration
1982	Phosphate ban called for in Parliament with a 3 year transition
1982-1983	Federal Commission on Water report that phosphates should not be used in detergents
1985	Federal ban
1986	Ban comes into effect

3.9.2 Surface water quality improvements

Lake Geneva

The first signs of eutrophication in lake Geneva were noticed in 1965, and particularly high levels occurred between 1974 and 1976 (CIPEL 2001). As can be seen in figure 3.10, from the late 1960s phosphorus concentrations had continued to rise before levelling off in the late 1970s (CIPEL 2001).

A decline in total phosphorus was observed between the late 1970s and 1995, from a high of around 0.9 mg/L to 0.4 mg/L in 1995. Since this date the concentration has remained relatively constant. This decline coincides with improved waste water treatment and the restrictions on the phosphate content of detergents from 1981 up to the total ban of 1986.

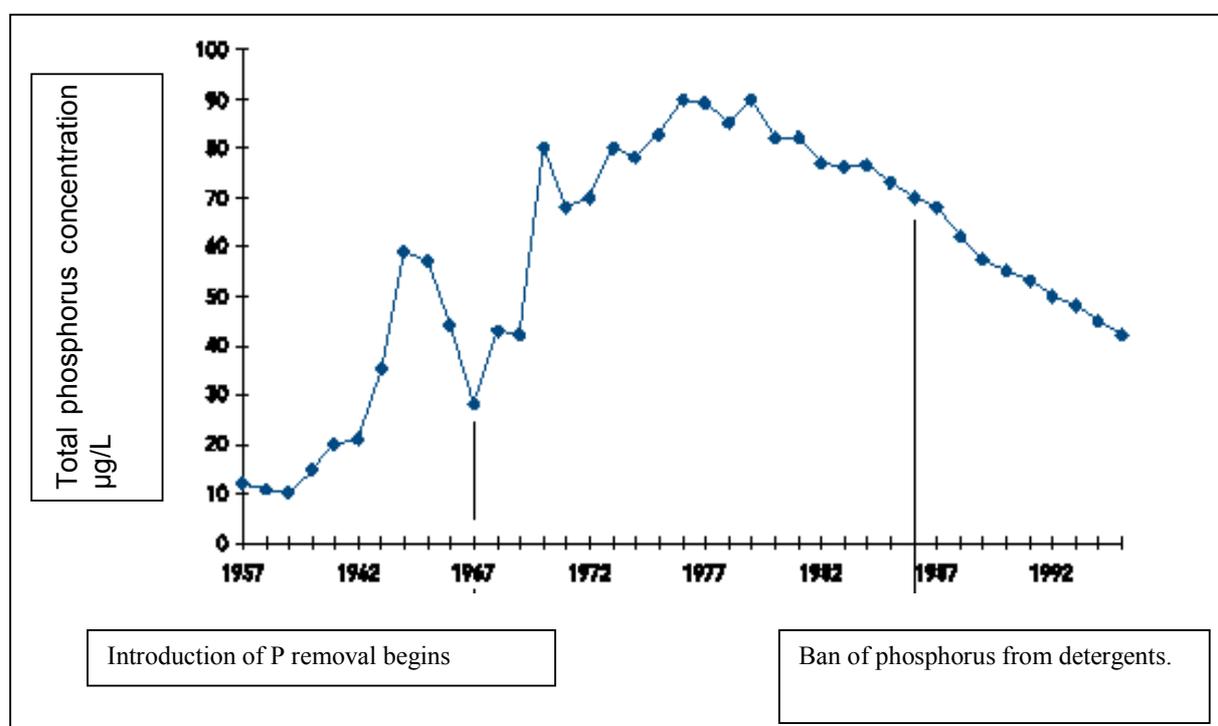


Figure 3.10 Total phosphorus concentration in Lake Geneva, 1957-1995

Despite of a clear reduction in P concentration, the overall biomass has not changed in the Lemman Lake. However, there has been a change in the species composition to reflect those more characteristic of an oligotrophic environment, indicating the trophic evolution of the lake. In particular there has summer blooms of cyanobacteria and dinophyceae have been replaced by zygothryceae and xanthophyceae. There is also some sign of presence of oligotrophic species such as the diatoms *synedra* and *cyclotella* (CIPEL 2001).

Until the end of the 1980s, the maximum chlorophyll a concentrations measured in May, June, July and August were located in the upper part of the euphotic layer (between the surface and

5 m deep). Since the 1990s, these maxima have been measured lower and up to 15 m showing a clear sinking of phytoplankton during the summer months.

Other water bodies

Some examples of other water bodies are given by Wehri (1997). The river Glatt shows a large reduction between 1985 and 1990 (the main sewage treatment works discharging upstream did not have nutrient removal at the time). By contrast sewage treatment at lake Zurich was more advanced at the time, and the reduction in P concentration is smaller. The reduction at lake Sempach between 1985 and 1990 is considerable (30%), but the relatively high 1995 concentration is due to animal husbandry on the Lucerne plateau.

Concentration mg P/l	River Glatt at Rheinsfelden	Lake Zurich	Lake Sempach
1985 (pre STPP ban)	0.07	0.06-0.07	0.17
1990 (post STPP ban)	>0.02	0.05-0.06	0.12
1995	0.02	0.03-0.04 ¹	0.06 ¹

Note 1. Predicted, not measured.

Phosphorus concentrations in Lake Murten illustrate a similar downward trend falling from over 0.14 mg/L in 1982 to just over 0.04 g/L by 1994. The combined effect of phosphate removal from wastewater and the banning of phosphate from detergents can also be seen in the Rhine at Basel. Phosphorus concentrations fell from approximately 0.17 mg/L of total phosphorus in 1980, to approximately 0.14 mg/L by 1985, with a further reduction to approximately 0.06 mg/L by the mid-nineties.

EAWAG² has estimated that the ban on STPP in detergents and improved sewage treatment have together resulted in a reduction of 2600 t P a⁻¹ in phosphorus entering surface waters in Switzerland from municipal wastewater, comparing 1980 with 1994. This represents nearly 60% of the 1980 load.

As a result of the STPP ban the requirement for chemical precipitation agents has been halved (Siegrist 1997). The total quantity of sludge produced has hardly changed, as the Zeolite A counteracts the reduced amount of phosphorus. This topic is considered in more detail in section 7.

The agricultural nature of the lake Sempach catchment also suggests that the lake has encountered a long history of enrichment from both diffuse and point sources. Therefore, it can be expected that Lake Sempach will require a longer period to recover from a reduction in external load, due to high internal phosphorus loading from the sediments.

The effects of the ban have in fact gone beyond the expectations of Swiss officials. In 1983 the Federal Office of the Environment concluded that the banning of phosphates from detergents would result in a reduction of 15 – 45%; by 1993 these expectations had been exceeded in 9 out of 10 Swiss lakes through a combination of the ban and improved wastewater treatment.

² Swiss Federal Institute for Environmental Science and Technology

3.9.3 Conclusions

The combination of improved waste water treatment and a reduction in phosphates in detergents have shown positive results for Swiss surface waters, as shown by the improved quality of Lake Geneva, Lake Murten and the Rhine River.

3.10 The USA

Legislative Background

Phosphorus use in the United States followed a similar pattern to other countries with use in fertiliser and detergents rising steadily from the 1950s and reaching a peak in 1970. The use of powdered washing formulations began in the late 1940s in the USA and rose rapidly so that by 1967, 220,000 tonnes were in use. Up to the late 1960s STPP use was as much as 60% by weight and phosphorus up to 15%.

The trend in the United States, as in Europe, has been to move towards liquid concentrates that use alternative builders that are not affected by hardness. In 1994 a voluntary industrial agreement was reached for the banning of phosphorus in domestic laundry detergent.

There are two elements to consider in relation to the downward trend in the levels of phosphorus discharged in municipal wastewater. The first is the national legislation of 1972. The 'Water Pollution Control Act (known as the Clean Water Act) led to a multi-billion dollar investment programme to up-date and improve the waste water treatment capabilities across the USA.

The second contributing factor in the decrease in phosphorus levels is state legislation, which also came to the fore in the early seventies with the introduction of bans on detergents using phosphorus.

Both the national and state responses were, in part, fuelled by public pressure to act following the identification of eutrophication processes occurring in many of the country's lakes (USGS 1999). Lake Erie and the other Great Lakes attracted national attention, and the USEPA began a nationwide survey in 1972. It was found that 10 to 20% of all lakes and reservoirs in the USA were eutrophic, and that phosphorus was the limiting nutrient in two thirds of the lakes studied. It was estimated that a phosphorus detergent ban by itself would improve the trophic status of one fifth of the lakes, and in the remainder additional P control measures would be needed.

Moves to regulate phosphorus use in detergents began in 1967 with the establishment of the Joint Industry-Government task Force on Eutrophication. There followed a call by Congress to end use of phosphorus use in detergents by 1972 but concerns regarding the safety of available substitutes (NTA and sodium carbonates, sodium silicates). In 1970 industry voluntarily agreed to limit phosphate in detergents to 8.7% by weight.

No federal legislation was introduced in the USA but individual states soon began to impose their own bans (table 3.20) and in 1971 5 cities in Illinois became the first to limit phosphates in detergents.

Table 3-20 Summary of USA policy development and legislation

Policy	Measures
Joint Industry-Government task Force on Eutrophication (1967)	There followed a call by Congress to end use of phosphorus use in detergents by 1972. Concerns regarding the safety of available substitutes (NTA and sodium carbonates, sodium silicates) were expressed.
First voluntary agreement (1970)	Industry voluntarily agreed to limit phosphate in detergents to 8.7% by weight
State bans begin (1971)	5 cities in Illinois became the first to limit phosphates in detergents. By 1995, 27 states have a ban in place. The majority of the bans apply to household products (New York also bans the use of phosphorus in commercial products) but do not apply to dishwasher. The bans take the form of a phosphorus limit as a percentage of phosphorus by weight, within the range of 0.5% and 8.7%
Clean Water Act (1972)	<p>Changed policy to effluent limitations, regulating the amount of pollutants being discharged from particular point sources.</p> <ul style="list-style-type: none"> • makes illegal the discharge of pollution without permit; • encouraged the use of the best achievable pollution control technology; and • provided billions of dollars for construction of sewage treatment plants. <p>Established guidelines for effluent</p> <p>Emphasis on control of point sources</p> <p>Between 1972 and 1991 \$200 billion was spent on the adapting of wastewater treatment plants and \$151 billion was spent on operating and maintenance. By 2000 the spending was up to \$600 billion on wastewater technologies.</p>
National Pollutant Discharge Elimination System (NPDES) as authorised by the Clean Water Act	Regulates point sources that discharge pollutants directly into waters. Industrial, municipal, and other facilities must obtain permits if their discharges go directly to surface waters. In most cases, the NPDES permit program is administered by authorised states.
Second voluntary agreement (1994)	Voluntary agreement with industry to remove phosphorus from domestic laundry detergent in 1994. The reason for the voluntary phase-out by industry was in part based on the costs involved of holding stocks of two different types of detergent across the country.
The Permit Compliance System (PCS) as authorised under the Clean Water Act (1997)	4.7% of industrial facilities were required to monitor for phosphorus and 1.7% had phosphorus limits. Of the 16,000 municipal wastewater treatment plants under the PCS (which covers 70% of the population) 15.3% were required to monitor for phosphorus, over half of which also have phosphorus limits. This covers 39% of the total municipal wastewater treatment plant discharge. The magnitude of limits, are for the main part within the range of 0.5mg/L to 1.5mg/L. Regional concentration around the East Coast and the Great Lakes.

Details of Policy Development

Clean Water Act

The Clean Water Act (CWA) created a federal program designed to achieve the goal of protecting and restoring the physical, chemical and biological integrity of waters in the USA. In addition to strengthening water quality standards system, this legislation:

- made illegal the discharge of pollution without permit;
- encouraged the use of the best achievable pollution control technology;
- provided billions of dollars for construction of sewage treatment plants; and
- Established guidelines for effluent

The 1972 act changed the thrust of enforcement from water quality standards, regulating the amount of pollutants in a given body of water, to effluent limitations, regulating the amount of pollutants being discharged from particular point sources. Under the Act states were required to report (biennially) on the quality of surface waters. The USEPA was further obliged to compile this data in to the National Water Quality Inventory Report, the first of which was published in 1974.

A major element of the Clean Water Act has been to achieve water improvement through the control of emissions from point sources. Between 1972 and 1991 \$200 billion was spent on upgrading wastewater treatment plants and \$151 billion was spent on operation and maintenance. The central goals of the Clean Water Act are “to restore and maintain the chemical, physical, and biological integrity of the nation’s waters.”

The \$200 million spent on wastewater treatment did not significantly increase the number of wastewater treatment plants, but it did increase the number of plants with secondary and tertiary treatment. Since the early 1970s, private and public sectors have spent more than \$500 billion on water-pollution control, much of which has been directed toward municipal and industrial point sources.

Investments under the Clean Water Act have seen the proportion of treatment plants with primary or no treatment fall from around 50% in 1968 to almost zero by 1996, while tertiary treatment had risen to around 25% of all plants by 1996. Currently, the treatment system in the United States provides 82% removal of BOD and TSS.

The investment programme under the Clean Water Act increased the number of treatment plants from 14,000 in 1958 to 17, 000 by 1996. The level of treatment was also significantly changed. Despite the large-scale investments in wastewater treatment plants only 7% of plants have tertiary treatment for the removal of phosphorus, hence there is still scope for further reductions in concentrations levels from point sources.

Other measures taken by the USA Environment Protection Agency include, the National Pollutant Discharge Elimination System (NPDES) permit program, (as authorised by the Clean Water Act). This program controls water pollution by regulating point sources that discharge pollutants directly into waters. Individual homes that are connected to a municipal

system do not need an NPDES permit; however, industrial, municipal and other facilities must obtain permits if their discharges go directly to surface waters. In most cases, the NPDES permit program is administered by authorised states.

One other aspect of the NPDES is the pre-treatment requirement. The General Pre-treatment regulations establish responsibilities of Federal, State, Local Government, industry and the public to implement Pre-treatment Standards to control pollutants from the industrial users which may pass through or interfere with POTW treatment processes or which may contaminate sewage sludge.

State Legislation

By 1995 27 States had implemented either complete or partial bans on the use of phosphate in laundry detergents. These are mainly those situated along the eastern-coast or around the Great Lakes. The most recent state to impose a ban was New Hampshire in 1995. This followed the voluntary agreement of industry to remove phosphorus from domestic laundry detergent in 1994. The reason for the voluntary phase-out by industry was in part based on the costs involved of holding duplicate inventories across the country.

Figure 3.11 shows there has been a steady increase in the number of state bans from the early 1970s with the last ban being in place put in place in 1995. The majority of the bans apply to household products (New York also bans the use of phosphorus in commercial products) but not apply to dishwasher detergents. The bans take the form of a phosphorus limit as a percentage of phosphorus by weight, within the range of 0.5 and 8.7 (table 3.21).

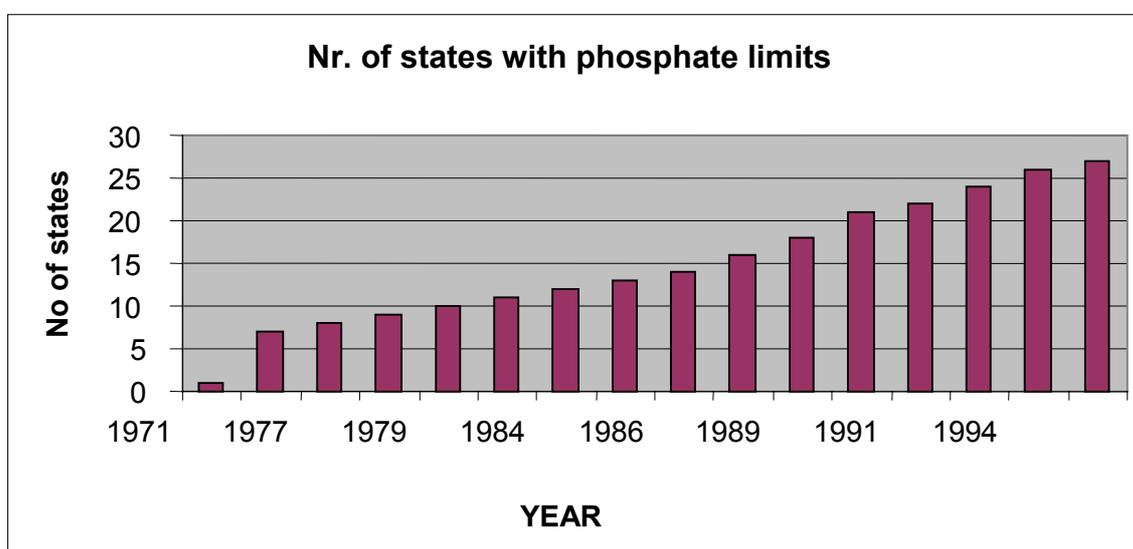


Figure 3.11 Phosphate limits in US States (1971-1995)

Table 3-21 USA state bans on STPP in detergents

state	Year of ban	Phosphorus limit by percentage of phosphorus by weight
Arkansas	94	0.5
Connecticut	72	8.7
District of Columbia	86	0.5
Florida	72	8.7
Georgia	89 (voluntary) 90 compulsory	0.5
Idaho	89	0.5
Indiana	72, 73	Limit reduced to 0.5 in 1973
Illinois	71-	Five cites 8.7 9 cities 0.5
Maine	72	0.5
Maryland	85	0.5
Massachusetts	94	0.5
Michigan	72	Limit reduced to 0.5 in 1977
Minnesota	77	0.5
Missouri		0.5
Montana	84	0.5
New Hampshire	85	0.5
New York	72, 73, 76	Limit reduced to 0.5 in 1973 and extended to commercial products in 1977
North Carolina	88	0.5
Ohio	88	0.5 (32 counties)
Oregon	92	0.5
Pennsylvania	90	0.5
Rhode Island	95	-
South Carolina	92	0.5
Texas	91	Austin and 3 cities only
Vermont	78	0.5
Virginia	80	0.5
Washington	90	0.5
Wisconsin	79, 82, 84	Ban lifted in 1982 and re-instated in 1984

Public Opinion

The involvement of the public in the push to reduce eutrophication in the USA has played a central part in the measures taken by both federal and state agencies. The evidence for this involvement comes from various areas including academia and pressure groups. For example the pressure group Environmental Defence Membership has been involved in campaigns relating to eutrophication since the early 1970s.

In 1972 EDF assembled an interdisciplinary team, which studied the Corps of Engineers' plans for the Tocks Island Dam on the Delaware River, and made a report to the Delaware River Basin Commission (DRBC). The report criticised the Corps' lack of scientific analysis in the areas of flood control, water supply, eutrophication, and economics.

In academia Joshua Lederberg, a microbiologist who received the Nobel Prize in 1958, was one of the many scientists voicing concern about the problem of eutrophication.

The Clean Water Act specifically provides for citizen participation in the enforcement of Federal standards. Private citizens may seek judicial relief against any polluter for violations of

an effluent standard or limitation, or administrative order issued under the Act. Citizens may also institute proceedings against the Administrator if he fails to perform an act required of him under the law.

Wastewater treatment

The Permit Compliance System (PCS) provides information on companies that have been issued permits under the NPDES to discharge wastewater into rivers.

Under this scheme industrial facilities as well as municipal wastewater treatment plants are required to monitor for phosphorus. This covers 39% of the total municipal wastewater treatment plant discharge. However, only half of the monitoring sites also have limits.

Phosphate limits are assigned a start date on the PCS database. Prior to 1973 few limits were in place. Between 1975 and 1995 limits were implemented at many sites, reaching a peak of 104 in 1986. The magnitude of limits, are for the main part within the range of 0.5 mg/L to 1.5 mg/L. One other aspect of the implementation of limits is the regional concentration around the East Coast and the Great Lakes with few limits being implemented west of the Mississippi River.

Summary of facilities in PCS data base with phosphorus monitoring requirements or limits

Type of facility	Total number of facilities	Facilities required to monitor for phosphates		Facilities with phosphate concentration limits	
		number of facilities	%	number of facilities	%
Municipal	15 939 ¹	2 437 ²	15.3	1 163	7.3
Industrial	50 599	2 379	4.7	877	1.7
Federal	1 119	110	9.8	50	4.5
Other	5 087	142	2.8	26	0.5
	72 744	5 068	7	2 116	2.9

Note 1: These treat sewage from 70% of the population.

Note 2: 39% of municipal wastewater discharged.

Nationally there is no historical database of phosphorus loadings from wastewater treatment plants. Trend analysis of concentrations of phosphorus from waste water treatment plants illustrates that between 1974-81 downward trends were seen in 50 locations mostly in the Great Lakes and Upper Mississippi region and mostly in point source dominated areas. Upward trends were indicated in 44 stations in which agricultural sources dominated while 288 stations showed no trend. Between 1982 and 1989 the number of stations displaying a downward trend rose to 90 (22%). The geographic spread of the stations with downward trends is scattered across the USA with the exception being the south-eastern region (USGS 1999).

Regional analysis shows a similar pattern. In the Mississippi River basin between 1974 and 1994, 25 out of 40 stations showed a downward trend in wastewater concentrations of phosphorus. The Gulf of Mexico showed 11 stations out of 37 showing a downward trend in data analysed between 1972 and 1992.

Phosphorus in surface waters

Under the Clean Water Act of 1972 states were required to report (biennially) on the quality of surface waters. The USEPA was further obliged to compile this data in to the National Water Quality Inventory Report, the first of which was published in 1974.

The first publication of the National Water Quality Inventory (1974) found that from the mid-1960s to the early 1970s total phosphorus concentrations had risen by 82% in 22 of the largest rivers and 57% of the assessed reaches exceeded the recommended total phosphorus level of 0.1 mg/L. For comparison, the USEPA STORET data for 1990-5 showed that in 32% of the hydrological units examined, phosphorus exceeded the recommended level in more than 50% of samples; in a further 44% of units, 10-50% of samples exceeded the recommended level.

The USGS conclude: "These data indicate that the potential for nutrient impairment in surface water remains substantial in the United States".

From Policy to Impacts - Specific Examples

By the 1980s sufficient data had been collected at specific sites to show considerable reductions of phosphorus concentrations could be achieved, through a combination of measures including wastewater plant upgrades, enhanced compliance with permits, and bans on phosphorus detergents.

- Lake Erie: annual municipal P load reduced from 14000 tonnes in 1972 to 2000 tonnes in 1990.
- Chesapeake Bay: point source P load reduced from 5100 tonnes in 1985 to 2500 tonnes in 1996.

In these two areas, the biological response has been monitored.

- Lake Erie. The central basin is critical because the eastern basin is sufficiently deep not to be threatened by oxygen depletion, while the western basin is shallow enough for complete mixing to occur. The summer average P concentration in the central basin reduced from 0.14 mg/L in 1968-72 to 0.08 mg/L in 1994-6 (Charlton et al 1999). "As a result, excessive growth of algae declined and the algal composition shifted toward more desirable ... species. However the problem of hypolimnetic oxygen depletion had not been ... reduced by as much as was expected" (USGS 1999). Charlton et al reported dissolved oxygen in late summer between 2 and 6 mg/L since 1993, except for 1998 when widespread concentrations below 1 mg/L were found.
- Chesapeake Bay: total P concentrations decreased by 16% between 1984 and 1992. "There has been no clear trend in oxygen levels in the bay, or in the health of bottom-dwelling organisms, but there are signs of improvement in the health and diversity of plankton communities..." (USGS 1999).

Sources of Phosphorus

One of the possible reasons for the persistence of eutrophication alongside falling levels of phosphorus from point sources is a rise in the use of phosphorus fertilisers, particularly in the 1980s.

Phosphorus use in fertiliser rose sharply from the 1950s through to the 1980s, peaking at 2.5 million tonnes. Use now appears to be holding at 2 millions tonnes per annum. Measurements of phosphate in soils between 1940 and 1980 were relatively static but recent years have seen a rise. In Wisconsin for example the average soil test level of phosphorus has risen from 34 mg/kg in 1967 to 48 mg/kg in 1990.

The problem of agricultural sources of phosphorus is further illustrated by data from the USEPA Clean Water Plan for 1998. This data found the main source of river and lake pollution by nutrients is now water run off from agriculture, indicating that the control of point sources combined with the state bans has had a major effect on phosphorus loadings from waste water sources. For estuaries, the major source of nutrients is still municipal waste water.

	% of those assessed that were impaired	Nutrients as cause (% of total impaired)	Main cause of impairment (% of cases of impairment)
Rivers	35%	29%	Agriculture (60%) Municipal point sources (35%)
Lakes	45%	44%	Agriculture (major) Municipal point sources (minor)
Estuaries	43%	23%	Municipal point sources (major) Agriculture (minor)

Source: USA National Water Quality Inventory, 1998.

An example of a catchment dominated by agricultural inputs is the upper Snake River Basin, in Idaho and Wyoming (Clark et al, 2000). The area of 90000 km² contains 400000 inhabitants. The northern and eastern parts are mountainous, and forested or used for non-intensive cattle grazing. In the Snake River plain are urban areas, fish farms and irrigated agriculture. The number of beef and dairy cattle has increased since 1980. It is in these areas that application of fertilisers and pesticides to land degrades river quality. However, urban wastewater discharges are not insignificant, and of the 27 sewage treatment works (that treat 180000m³/d) only one has phosphorus removal, that treats 9000m³/d.

Conclusions

A combination of measures have led to a lowering of phosphorus levels in the water ways of the USA these policies include:

- political and public pressure on industry leading to voluntary banning of phosphorus in domestic laundry detergents
- large-scale investments that were triggered by the Clean Water Act of 1972
- implementation of phosphorus limits on waste water.

The Lake Erie program is a prime example of the US policy of tackling eutrophication through the reducing the loadings of phosphorus entering water via waste water by improvement of wastewater treatment and the application of bans of phosphorus in detergents.

The continued level of eutrophication reported in the National Water Quality Inventory, particularly in lakes across the USA may be explained by several phenomena. Firstly, the increased use of phosphorus fertilisers, resulting in the transport of phosphorus to waterways via run-off. Secondly, only 7% of sewage treatment plants have phosphorus removal. Thirdly, phosphorus detergents are still used in dishwasher and non-domestic applications. Finally, there may be a tendency to concentrate on lakes experiencing problems than on a representative sample. Hence the results are skewed towards problem areas.

Assessing the relative importance of each of the policies mentioned above is problematic without further corroborative evidence regarding the timing of the reductions in phosphorus levels. In particular further data are required to separate out the impact of the Clean Water Act of 1972 and the voluntary agreement banning the use of phosphorus from domestic laundry detergents in 1994.

Nevertheless, the current data indicate that this mixture of policies have resulted in a significant fall in phosphorus concentrations from the peak of 1970.

The USA offers an important case study of how public pressure, government, and state legislation can combine to stimulate research looking for alternatives to well established products, resulting in the adoption of less pollutant products at a low cost to industry.

4. DETERGENT ECOLABEL SCHEMES

Pan-European schemes aimed at minimising the effect of detergents are also in place. Two of the most prominent are the Eco-label and the Nordic White Swan. The aims of both are similar and encompass encouraging business to market greener products. The twin goals of the schemes are to provide producers with the necessary information to take advantages of this strategy, and to enable consumers to make informed decisions regarding the environmental impact of products.

The Nordic environmental label is a neutral, independent label which guarantees a certain environmental standard and works in close co-operation with the eco-label scheme and is run through the competent bodies as nominated by the members, (Sweden, Finland, Denmark, Iceland and Norway).

The label helps consumers to identify the products that cause the least damage to the environment amongst those in the market. As a result, manufacturers are stimulated to develop products and production processes better for the environment.

Under the EU eco-label scheme (as laid down in the new Regulation (EC) No 1980/2000) each Member State has established a Competent Body, your first point of contact, to run the Scheme at the national level. At the European level the Scheme is run by the EU Eco-labelling Board (EUEB), this consists of the Competent Bodies and the different non-governmental interest groups. Furthermore, other countries outside of the EU and EEA follow our work as observers to the scheme.

Only products which satisfy strict environmental requirements on the basis of objective assessments are allowed to display the either of the labels.

The Nordic Swan criteria for “all purpose cleaners” allow :

0.2 g P (phosphorus) per recommended dose per litre (this means per litre after dilution according to manufacturer’s recommendation)

The Nordic Swan criteria for “sanitary” cleaners allow :

- 2 g per 100g of P (phosphorus)

These 0.2 g/l and the 2 g/100 g limits on P correspond to approximately 0.008% and 8% STPP.

The European Union’s official “Ecolabel” scheme :

Laundry detergents : (1999/476/EC)

- 30 g STPP within a maximum 110g total chemicals/wash = 27% STPP

Automatic dishwasher detergents : (1999/427/EC)

- 10 g STPP within a maximum 22.5 g total chemicals/wash = 44% STPP

All purpose and toilet cleaners, hand dishwasher detergents are currently in the process of criteria definition.

The current eco-label-criteria also promote consumer information about 'dosage' and 'low temperature washing' only. Thus it is suggested that further use instructions are added to reduce environmental impact, these include:

- Pre-sort laundry (by colour, degree of soiling, type of fibres)
- Treat specific soils (ink, fruit, etc.) prior to wash
- Wash with full loads
- Avoid pre-washing
- Avoid overdosing
- Prefer low temperature washing cycles

The Nordic environmental label is the official ecolabel in Norway, Sweden, Denmark, Finland and Iceland.

5. THE PHOSPHATE & ZEOLITE INDUSTRIES IN EUROPE

5.1 STPP Production

There are two distinct parts to the phosphate industry in Europe; the fertiliser industry and the chemical industry. Both use the same basic sources of raw materials but process and refine them in different ways. There is very little interaction between the two parts.

The fertiliser industry requires lower levels of purity than the chemical industry, and uses the majority of the imported phosphorus rock or phosphoric acid. The chemical industry refines phosphate rock or green phosphoric acid prepared at the original sources, to products which are used in industries as diverse as food and drinks, metals, pharmaceuticals and biotechnology.

While this study considers briefly effects of fertilisers on the environment ([section 5.2](#)), this section deals specifically with production of the ingredient sodium tripolyphosphate (STPP), which is used as the builder in detergents that contain phosphorus. STPP is the major non-fertiliser phosphorus product in current chemical production, and is regarded by much of the industry as the bulk product that supports production of other fine chemical or food grade phosphate products. Industry representatives have stated that, in the absence of production of STPP for detergents, their individual facilities would be unlikely to be able to continue producing other phosphate products at prices competitive with other world sources. In support of this, the data in [table 5.2](#) suggest that 60% of the non-fertiliser use of phosphates is in detergents, and that European manufacture is on a relatively small scale.

In Europe, the USA and other countries where the environment is a major issue, the use of STPP in detergents has either been discontinued or has fallen. In other countries such as China, Russia and in Latin America, the use of detergents is increasing, and there is little tendency to try to minimise the use of STPP. The international nature of the chemical industry suggests that European manufacturing facilities may be vulnerable to external competition, with the possibility of relocation to China (for example) where there are lower costs and environmental constraints. This prospect would be made more likely by a ban on STPP in detergents.

The world annual phosphate production (P_2O_5 equivalent) is in the region of 45 million tonnes per year ([table 5.1](#)).

Table 5-1 World production of phosphate, 1995 - 1999

	Production (million tonnes)				
	1995	1996	1997	1998	1999
World rock	131	135	143	144	141
Phosphate (P₂O₅)					
World	40.8	41.8	45.1	44.9	44.1
US	12.8	13.3	13.3	12.9	11.8
Morocco	6.4	6.6	7.8	7.9	7.9
China	5.8	6.3	7.5	7.5	7.5
Russia	3.1	2.9	3.3	3.3	3.7
Tunisia	2.2	2.2	2.1	2.4	2.4
Jordan	1.7	1.8	1.9	2.0	2.0
Brazil	1.4	1.4	1.5	1.6	1.5
Israel	1.3	1.2	1.3	1.3	1.3
South Africa	1.1	1.0	1.1	1.1	1.1
Rest of the World	5.1	5.2	5.2	5.1	5.0
Total European	0.36	0.35	0.36	0.36	0.36
% of world	0.87%	0.85%	0.79%	0.81%	0.81%
Finland	0.244	0.246	0.254	0.260	0.260
France	0.018	0.014	0.008	0.008	0.008
Germany	0.019	0.019	0.019	0.019	0.020
Luxembourg	0.075	0.075	0.075	0.075	0.070

Source: US Geological Survey

Some recent values of phosphate raw materials are these:

- Value of phosphate rock in USA, \$26.39 per tonne in 2000; cf \$30.43/tonne in 1999.
- Current value of P₂O₅ from rock, USA \$160/tonne.
- North African Di Ammonium Phosphate \$165/tonne in Europe.

Table 5-2 World uses of phosphate

	Million tonnes P ₂ O ₅ equivalent, 1995						Total	Total
	W Europe	CEE & FSU	USA	Other developed	Developing	Total		
Mineral fertilisers	3.5	2	4	2.5	19	31	80%	
Detergents	0.4					4.7	12%	
Animal feeds						2	5%	
Specialities ¹						1	3%	

Note 1. Food, metals, pharmaceuticals

5.2 Phosphate rock extraction and phosphate manufacturing processes

Rocks that contain phosphates are distributed throughout the world, in two main types; igneous and sedimentary deposits. Phosphorus in igneous rocks is abundant and widespread. The apatite family of phosphorus minerals (for example, fluorapatite, Ca₅(PO₄)₃F, or

hydroxylapatite, $\text{Ca}_5(\text{PO}_4)_3(\text{OH})$) is the group used for industrial extraction from igneous rocks. The largest mined source of apatite is on the Kola peninsula, in the Russian arctic. Most igneous rock deposits contain relatively low concentrations of phosphate.

The Sedimentary deposits (amorphous phosphate rock, or phosphorite) were formed from sources that include marine invertebrate shells, vertebrate bones and excrement. 80% of world phosphate production is derived from sedimentary deposits. These are also forms of apatite, in mixtures with quartz, calcite, dolomite, clay and iron oxide.

Minor sources, about 0.25% of world production, include process slag and guano.

Although crystalline apatite deposits can be concentrated by flotation to achieve 88% BPL (Bone phosphate of lime, equivalent to $\text{Ca}_3(\text{PO}_4)_2$) it is not always competitive with phosphorite (at 60% -75% BPL), as the crystalline apatite requires a three times longer reaction time with sulphuric acid than the porous phosphorite for conversion to super phosphate fertilisers.

The phosphate content in rocks currently mined ranges from 40% down to 5%. Processing removes bulk impurities and increases the apatite content generally by 1½ to 9 times. After processing, phosphate rock ranges from 32% up to 42% P_2O_5 .

A smaller percentage of high grade rock is used now compared to 30 years ago. Reserves of economically extractable rock have been assessed by a variety of organisations, and are in the region of 100 times the rate of current world annual extraction. Thus the US Geological Survey summarises annual extraction rates of in the region of 140 million tonnes of rock per year, with available known reserves of 12 to 30 billion tonnes. These are exploitable at costs of less than \$35/tonne. Potential reserves are more than twice as large; these reserves are exploitable at less than \$100/tonne (US Geological Survey, 1999; Steen, 1998). Total production costs including mine and mill operating costs, plus transport, taxes and investment returns are estimated at \$25-\$70/tonne of processed rock, with North African supplies in the region of \$35-\$45/tonne (Steen, 1998).

Heavy metal contaminants are present at higher levels in sedimentary rocks than in igneous rock. A particular issue is cadmium contamination, and for this reason limits have been applied to the cadmium content of fertiliser in some European countries. However, the absorbant nature of calcium apatites has resulted in phosphorite rocks absorbing many other metal contaminants from sea water, such as uranium, nickel, chromium, copper and zinc. Thus residues from processing the phosphate rocks may be treated as hazardous wastes, although in some cases building material products are made with the residues (personal communications from ThermPhos International BV and Chris Thornton, CEEP).

The majority of phosphate rock extraction is used for preparation of soil fertilisers. Phosphates for fertilisers require only limited purification beyond extraction from the phosphate rock, although there are limitations on the amount of contaminants that may be included. In particular there may be a need to remove fluorine and reduce heavy metal contaminants, which can be achieved by mixing processed rock with purified materials.

The use of phosphorus to produce chemicals for detergents, feed additives, and other special applications, requires greater purity.

5.3 Phosphoric acid manufacturing processes

Two main methods are used for extraction and purification of phosphorus or phosphorus salts from phosphate rock. The thermal process uses high temperature reducing conditions to prepare an elemental phosphorus vapour from phosphate rock. Wet processes use acidic extraction to form phosphoric acid. The thermal route produces the purest initial product, but is energy intensive and subject to energy and value losses from contaminants such as iron.

Most phosphorus is now extracted using the wet route, although in Europe the thermal route is a significant (20%) fraction of production capacities.

Thermal route

The rock phosphate is mixed with clay in a ball mill to form pellets (10 mm diameter) and baked in a carbon monoxide atmosphere to remove fluorine, and to form a dry compound. The dried pellets are added to an electric furnace together with coke and gravel. The furnace operates at 1200°C to 1500°C.



rock phosphate + sand + coke → calcium silicate + phosphorus + carbon monoxide

The phosphorus vapour is condensed and stored under water. Other metals, including zinc and cadmium, are also present in the vapour and dust, and are separated using cyclones or precipitators.

Some of the carbon monoxide product is recycled to the pellet sintering oven, and any remaining can be burned for generation of heat or power. The molten calcium silicate slag is tapped from the base of the furnace. The slag contains the majority of the metal impurities, including the radioactive elements, and so is mildly radioactive. As such, it can only be used for construction materials that are isolated from human exposure.

The phosphorus is burnt in air to form phosphoric acid in a highly exothermic reaction. The phosphoric acid produced is very pure, with only arsenic as a potentially significant contaminant.

Iron in the rock mixture is a significant problem, causing loss of energy and a reduced amount of phosphorus product as ferrophosphorus (FeP) is produced instead of phosphorus. Iron must be at less than 1% in the component materials for the thermal process to be economic.

Wet route

Finely ground phosphate rock is mixed with either sulphuric acid or hydrochloric acid. The hydrochloric acid route is used for only a small proportion of phosphate production.

The reaction with sulphuric acid produces phosphoric acid, and the process is controlled to encourage formation of calcium sulphate crystals (gypsum) which settle or filter out. After washing the gypsum is waste, as it contains impurities including radioactive elements. This 'beneficiation' process is now generally used at the site of extraction.

Most imports to Europe of phosphorus raw materials for use in non-fertiliser applications are in the form of the crude phosphoric acid rather than phosphate rock.

The phosphoric acid may be used directly in production of fertilisers, or further purified in cross-stream extraction systems to prepare high purity phosphoric acid as the source for other phosphorus products.

The hydrochloric acid route dissolves the calcium phosphate, to form a solution of monocalcium phosphate, which is then extracted by crystallisation, producing high grade mono and di calcium phosphates, with impurities remaining either in the initial acid extraction residue, or in the separated liquor.

Other variants of the basic sulphuric acid route produce various grades of phosphoric acid by using a variety of organic extractions and evaporations to purify an increase the product strength.

To achieve phosphoric acid suitable for detergent phosphate preparation generally requires no more than the sulphuric acid route, followed by removal of residual fluoride (as insoluble Na_2SiF_6), iron, aluminium, arsenic and other metallic impurities.

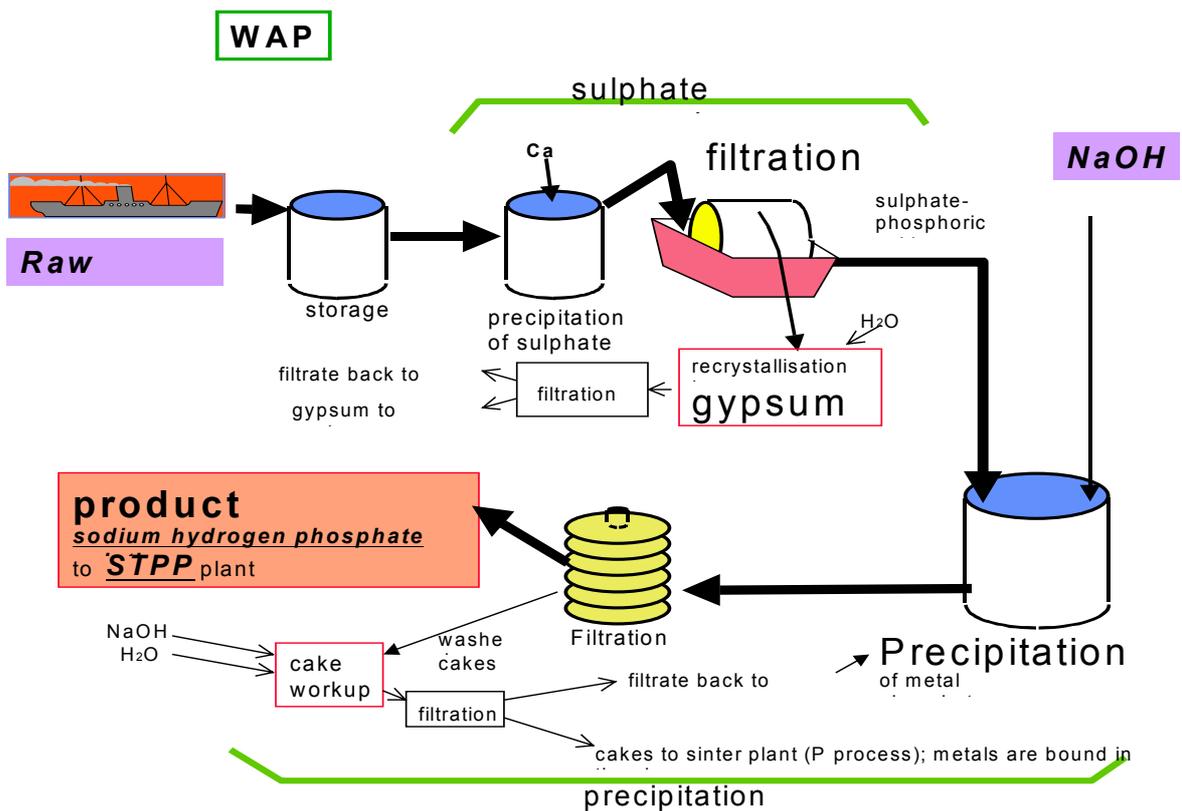


Figure 5.1 Crude acid purification³

³ Process at Thermphos bv, Vlissingen

5.4 Manufacture of Sodium Tripolyphosphate

Sodium tripolyphosphate, STPP, $\text{Na}_5\text{P}_3\text{O}_{10}$, is the main phosphate present in detergents. It is prepared from phosphoric acid by neutralisation with soda ash (sodium oxide) forming sodium hydrogen phosphates (see Figure 5.1). A powdered mixture of disodium hydrogen phosphate, and sodium dihydrogen phosphate is heated to 500°C to 550°C to produce the stable form of STPP (see Figure 5.2).

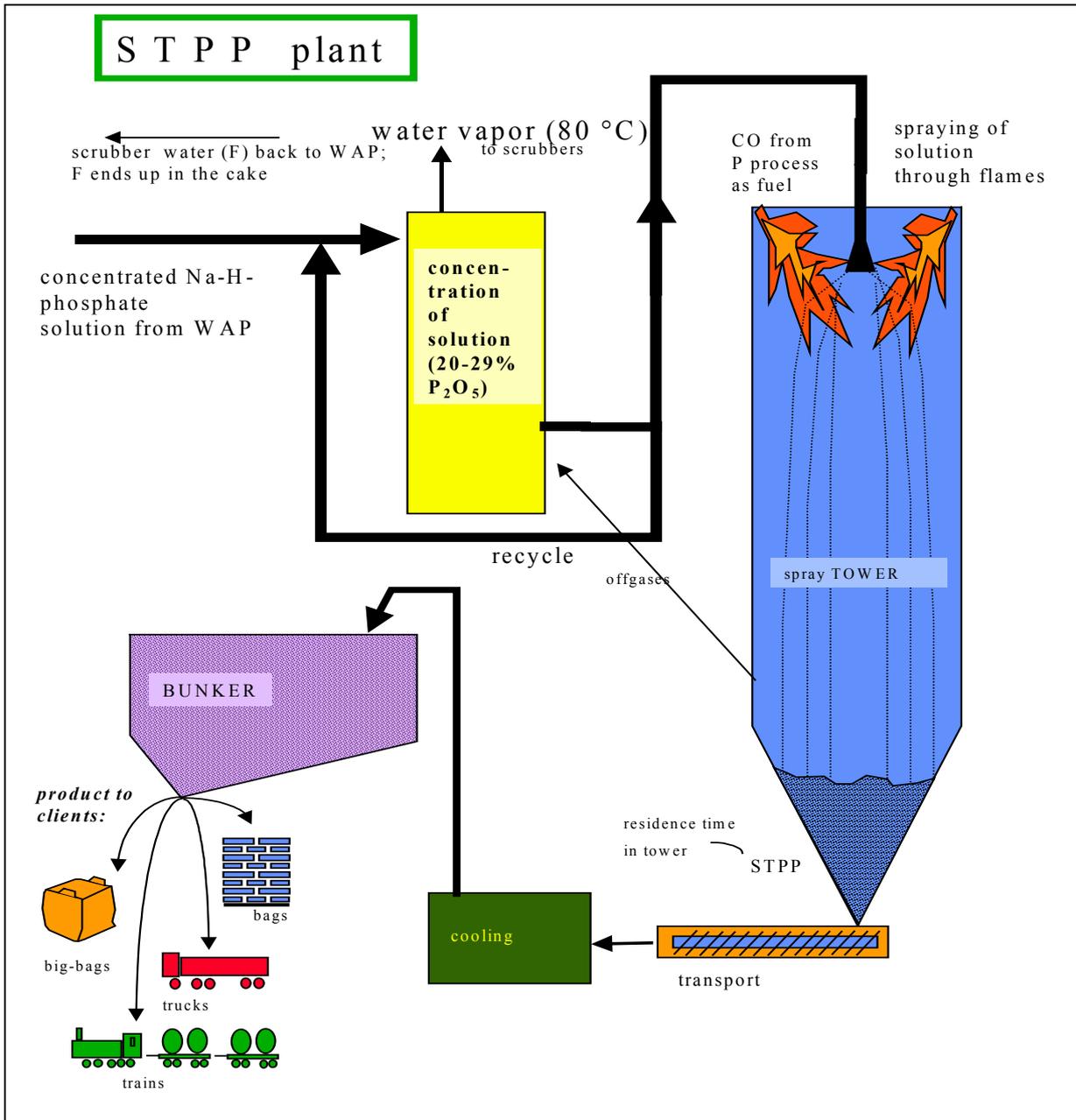


Figure 5.2 STPP production⁴

⁴ Process at Thermphos, Vlissingen

5.5 European STPP manufacturers

There are six chemicals businesses in Europe that manufacture phosphorus products including STPP (table 5.3). The businesses are based in Europe but have international connections as subsidiaries and associated businesses. Rhodia's UK phosphate production plant at Whitehaven has recently closed.

Table 5-3 European STPP manufacturers

Company name	Location	Numbers employed		% of the available market (EC document)	Maximum capacity (Kt pa gross)	Value (millions euro)
		General	STPP			
Rhodia	France, UK			30-40	?290	
Prayon	Belgium		100	5-15	?100	
FMC	Spain			10-20	?120	
Thermphos Kemira	Netherlands	400	400	5-25	180-(275)	
BK Giulini	Germany			0-10	?50	
CF Budenheim	Germany			0-10	?50	
CEFIC total (1997)			1000- 1500		820	430

The manufacture of STPP for detergents follows directly from the production of high quality phosphoric acid, which also provides for production of food grade STPP, and ammonium phosphates. Phosphoric acid is used as the base for manufacture of a wide variety of other phosphorus containing products. Examples of the areas in which the products are used are shown in the following table 5.4. Estimates of the total quantities produced for detergents are given in table 5.5.

Table 5-4 Examples of products that contain phosphorus

Industrial cleaning	Car wash, Food premises, other cleaning
Animal food additives	
Human food preparation and additives	Bakery, meats, cheeses, preservatives and supplements; emulsifiers, stabilisers, buffers, nutrients
Drinks additives	Acidulants ('Colas'), flavour enhancement, nutrients, clarifiers
Toiletry products	Dentifrice, other cleansers
Pharmaceuticals	Intermediates; supplements and buffers
Biotechnology	Analytical chemicals
Biocides	Organo-phosphates
Water and wastewater industries	
Metals industries	Surface cleaning, treatments, anticorrosion agents; extraction and refining
Textile production, washing	
Paint manufacture	
Flame retardants	
Oil industry	Lubricant additives
Ceramics, clay	
Paper industry	

Table 5-5 Estimates of detergent builder use in Europe

Quantity	Estimated quantity (000 tonnes/year)	Source of estimate
Total builder use	1036	25% of mass of laundry & dishwasher detergents
	1050	Zeolite manufacturers
Zeolite use	630	Zeolite manufacturers – 60% market penetration
	682	Detergent consumption by country
STPP use ¹	520	One STPP manufacturer's estimate
	420	40% of market penetration
	354 ¹	Detergent consumption by country

Note 1. This estimate is derived from the same source as [table 2.5](#), but with other non-EU European countries included, in particular Romania and Bulgaria where STPP use is high.

5.6 Zeolite A manufacturers in Europe

In Europe the usage of detergent Zeolites in 2000 was estimated by the trade association Zeodet to be 650000 tonnes (cf. table 5.5). This is probably a significant proportion of world wide use in detergents; Hauthal (1996) states that in 1994 slightly over 1 million tonnes of detergent grade Zeolite was produced worldwide, while Kemezis (1999) gives a lower figure of 850000 tonnes, of which 350000 tonnes was for the USA. The actual current figure may be higher, given that Zeolites are used in the USA, Japan, Canada as well as in Europe.

The main European manufacturers are:

<i>Name</i>	<i>Location of production plant</i>
Magyar Aluminium	Hungary
Sasol SpA	Italy
INEOS Silicas	UK
FMC Foret	Spain
Henkel KgaA	Germany
Industrias Quimicas del Ebro	Spain
Zeoline	Belgium
Silkem	Slovenia
PQ Europe	Netherlands

Roskill (1998) indicates that world-wide utilisation of detergent grade Zeolite manufacturing capacity is less than 60%. Utilisation in the USA is higher (80%), but in Europe it is similar to the world-wide figure.

5.7 Conclusions

World wide, STPP is used as a detergent builder more than Zeolites: 4.7 million tonnes compared to 1 million tonnes or somewhat more. Zeolites are used in the USA, Canada, Japan and much of the EU, while STPP is used in China and India.

The STPP production capacity located in the EU is relatively small compared to the rest of the world (<10%). Detergents represent a high proportion of STPP production, and if STPP use in detergents were banned in the EU, it is possible that production in Europe would cease.

Detergent Zeolite production in Europe (Hungary as well as the EU) is a relatively high proportion of world production, perhaps 50%.

There is currently spare production capacity in Europe for both STPP and Zeolite A. Any trend away from STPP and towards Zeolite use would have an adverse effect on the STPP producers; the increased demand for Zeolite A could probably be met without major investment in production capacity.

6. DISCHARGES OF PHOSPHORUS TO SURFACE WATERS

In this section, estimates of the quantities of phosphorus discharged to surface waters in municipal wastewater from households are presented. The current situation is compared with what would be expected to apply if:

- i) STPP were banned from detergents throughout the EU and accession states,
- ii) The Urban Wastewater Treatment Directive were implemented fully in the EU and accession states,
- iii) Both i) and ii).

The estimates are put in the context of discharges from industries and agriculture.

6.1 Industrial discharges of phosphorus

Industrial sources of phosphorus are comparatively minor in overall importance (table B.4), although they can be locally important. They are unlikely to increase, given the controls of industrial discharges that apply in the EU member states.

6.2 Agricultural inputs of phosphorus

Agricultural activity is a major source of phosphorus in surface water (table B.4 and appendix B generally). Runoff coefficients have been calculated for different countries and range from 0.2 to 1.4 kg P/ha/year for agricultural land (EEA 2001, chapter 14).

Unlike nitrogen, phosphorus applied as fertiliser is not very soluble. Only part is used by plants, and much of the remainder accumulates in soil. Figures for Switzerland (1994) illustrate the point (table 6.1). To control this accumulation, the practice of limiting the quantity of phosphorus that may be applied to soils where it is already available has been introduced (as in the UK Code of Good Agricultural Practice, Edge 2001).

Table 6-1 Phosphorus flows – agriculture Switzerland 1994

In	000 tonnes P per year	Out	000 tonnes P per year
Commercial fertiliser	13	Food	7
Compost, sewage sludge	3	Water environment	2
Animal feed	6		
Atmosphere	1		
Total	23	Total	9
Accumulation in soils			14

The majority of phosphorus entering surface water from diffuse sources is believed to come from erosion (62%) and surface runoff (24%) (UBA Deutschland 2001). It follows that phosphorus inputs are unlikely to decrease in the foreseeable future, and may increase even if fertiliser use falls.

Estimates of total inputs are available for a limited number of areas (table B.4). In primarily agricultural areas, or where the standard of municipal wastewater treatment is high, agriculture can be the largest single source of phosphorus. Any credible extrapolation of these estimates to the whole of Europe, or forward in time, would be beyond the scope of this report.

6.3 Municipal wastewater

The calculations presented here are for the domestic part of urban wastewater. Industrial discharges to urban sewer systems are excluded (but are believed to be small). Population trends have been ignored.

6.3.1 Detergent use

The quantities of detergent used per person are shown in table 6.2. Differences may be related to:

- Type of detergent used, e.g. traditional powder, compact powder, tablet (section 2.2);
- Water hardness – the harder the water the more detergent is needed;
- Clothes and dishwashing practice – Portugal, Greece, Hungary and the Czech Republic all have low detergent use, and it may be that this will rise as dishwasher and washing machine (?) ownership rises.

Factors that may contribute to a downward trend in detergent use are improved machine design and a change to more economical forms of detergent. In developing future scenarios, it has been assumed that future laundry detergent use will be 90% of the current EU average, or 6.4 kg/person/year. This corresponds to the AISE target of a 10% reduction. For automatic dishwashers, it has been assumed that ownership will rise throughout Europe, and that detergent use will reach 1.6 kg/person/day throughout Europe. This represents an increase in some countries.

One possible way of reducing the quantity of detergent used would be to adjust the detergent formulation to suit the hardness of the local water supply. This might be possible in Scandinavia, where water supplies are soft. Elsewhere in Europe both soft and hard waters are supplied in the same country, and the practical issues associated with distribution of the different formulations would need to be considered.

Table 6-2 Per capita detergent use

Country	Population (2000)	Detergent use (1998)		Detergent use	
		Laundry	Automatic dishwasher	Laundry	Automatic dishwasher
	millions	kilotonnes	kilotonnes	kg/head/year	kg/head/year
Austria	8.1	59	13	7.3	1.6
Belgium	10.2	78	15	7.6	1.5
Denmark	5.3	31	10	5.8	1.9
Finland	5.1	32	7	6.3	1.4
France	58.4	450	168	7.7	2.9
Germany	81.9	490	158	6.0	1.9
Greece	10.5	62	9	5.9	0.9
Ireland	3.6	41	3	11.4	0.8
Italy	57.3	415	36	7.2	0.6
Luxembourg	0.4				
Netherlands	15.5	100	21	6.5	1.4
Portugal	9.9	43	2	4.3	0.2
Spain	39.3	241	25	6.1	0.6
Sweden	8.8	44	9	5.0	1.0
UK	58.8	573	85	9.7	1.4
Total EU	373.1	2659	561	7.1	
Hungary	10.2	40	1	3.9	0.1
Czech Rep.	10.3	17	0	1.7	0.0
Poland	38.6	372	6	9.6	0.2
Total Acc'n States	59.1	429	7		
Total	432.2	3088	568		
Assumed long term values				90% of EU mean 6.4	4 th largest value 1.6

6.3.2 Discharges of phosphorus

Two sets of calculations have been done: by country and by major river catchment.

By country

For each country, the current municipal wastewater treatment situation has been summarised (table C.1). Data for this has been taken from various sources, that are noted at the foot of the table.

Country populations have been divided into village/town/city size bands (table C.2). These are based on data purchased from HarperCollins (Bartholemew Maps). Dispersed populations have been calculated as the difference between the total country population and that of the urban centres.

Assumptions about the proportion or concentration of phosphorus discharged to the water environment in treated wastewater are shown in table C.3.

Estimates of the standard of wastewater treatment that will apply after full implementation of the UWWTD are shown in table C.4. In countries where nutrient removal has been implemented for population centres below 10000, as well as above, the existing situation has been assumed to continue.

In deriving the estimates, assumptions have been made about the proportions of the population in some countries who live in sensitive areas. These are shown in table C.4.

The estimated quantities of phosphorus discharged are shown in table C.5. They are illustrated in figures 6.1 to 6.5 for 5 of the countries with major phosphorus discharges.

The figures confirm:

- Both wastewater treatment improvements and banning STPP from detergents reduce phosphorus discharges significantly.
- Their combined effect is less than the sum of the individual effects.

Significant quantities of phosphorus would still be discharged to surface waters, even after full implementation of the UWWTD, because:

- All the countries illustrated except Poland have significant non-sensitive areas,
- Dispersed populations are assumed to discharge 50% of phosphorus to the water environment,
- 60% of phosphorus from population centres up to 10000 enters the water environment.

Estimates of the phosphorus loads that would enter sensitive waters under different scenarios are shown in [table 6.3](#).

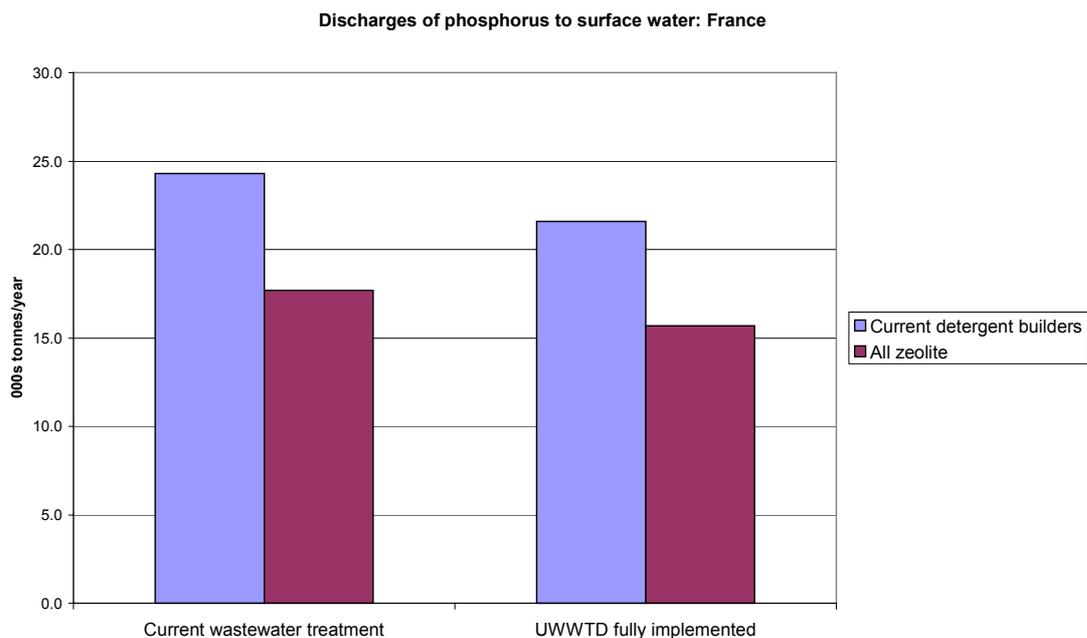


Figure 6.1 Discharges of phosphorus to surface water: France

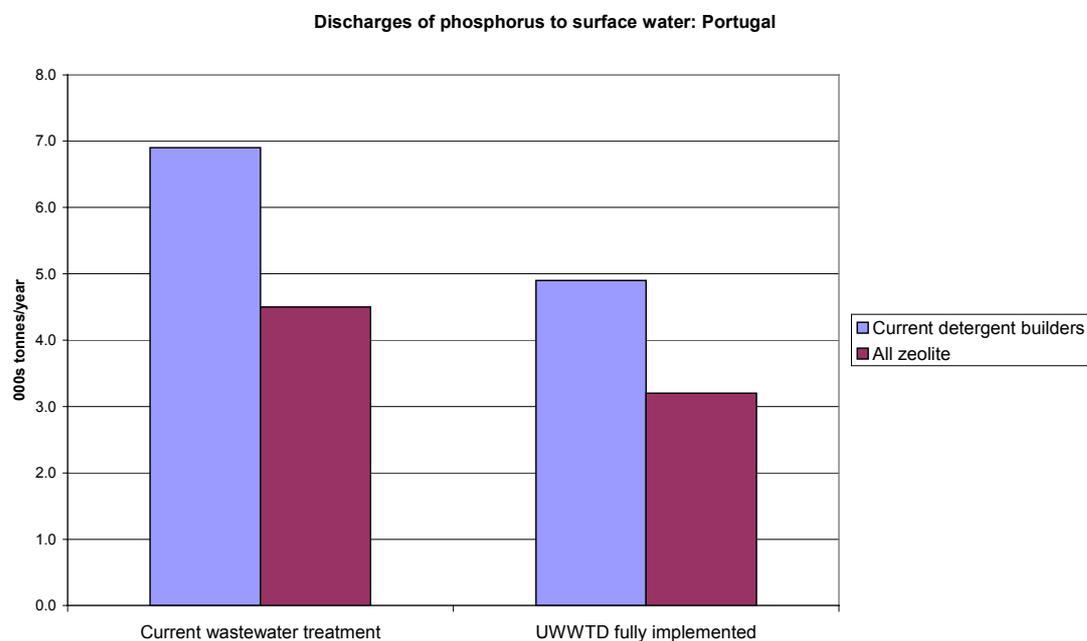


Figure 6.2 Discharges of phosphorus to surface water: Portugal

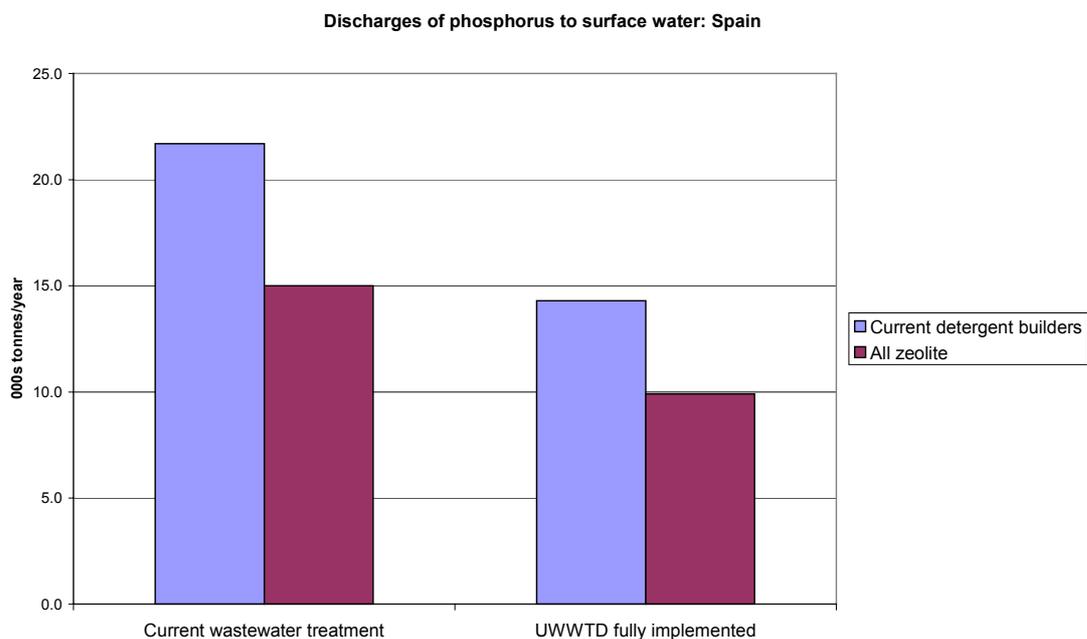


Figure 6.3 Discharges of phosphorus to surface water: Spain

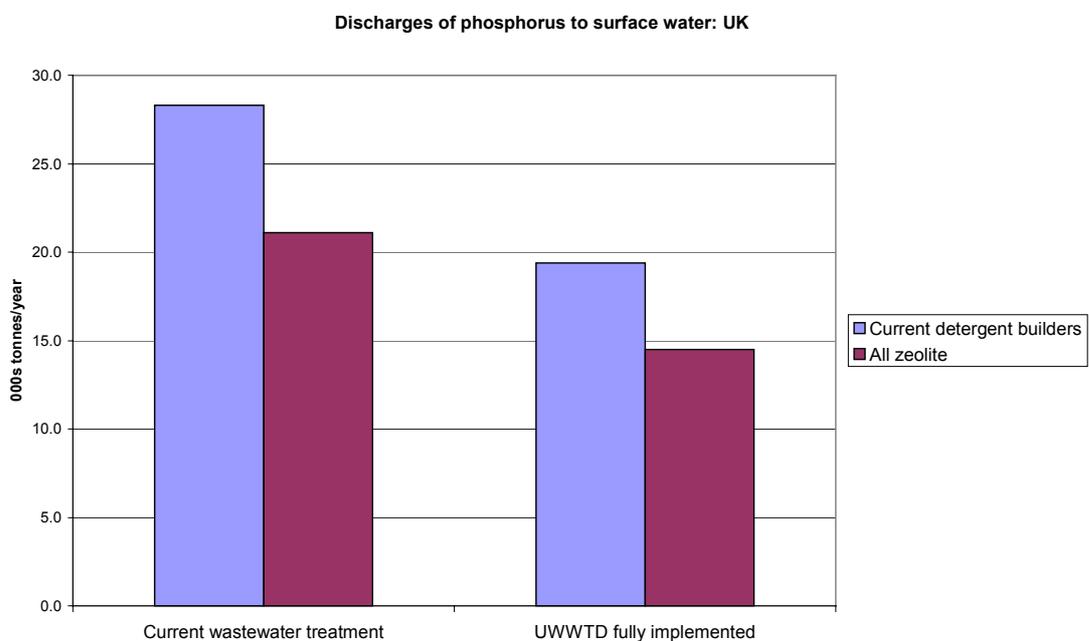


Figure 6.4 Discharges of phosphorus to surface water: UK

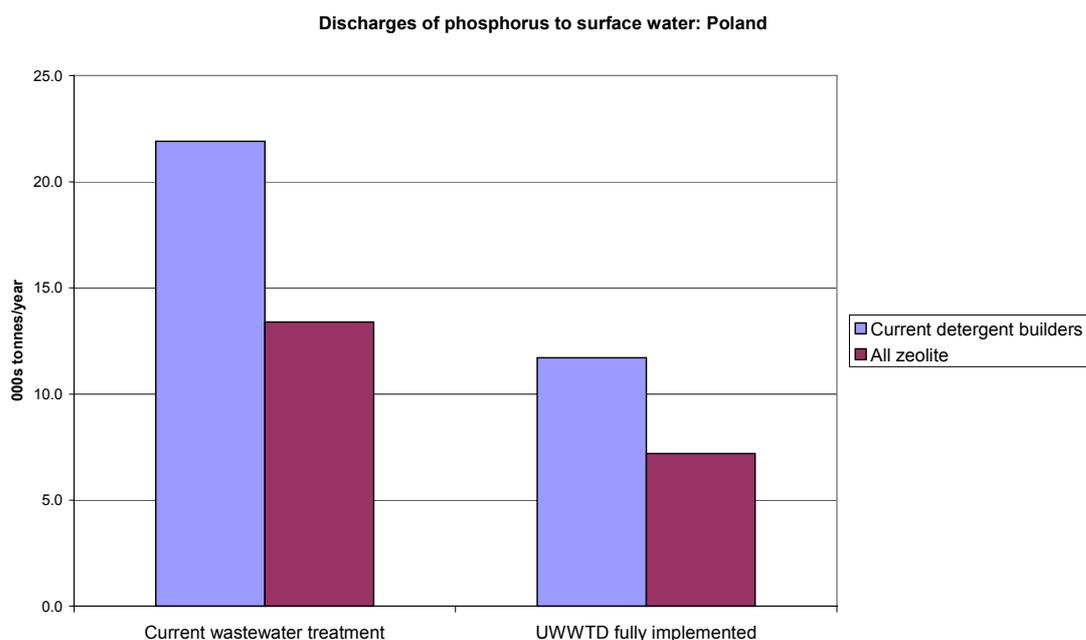


Figure 6.5 Discharges of phosphorus to surface water: Poland

Table 6-3 Estimates of phosphorus discharged to sensitive areas

Country	Assumed % population in sensitive areas	After UWWTD implementation	
		Current detergent builders	100% zeolite
		000 tonnes/year	000 tonnes/year
France	70%	14.9	11.4
Portugal	20%	0.8	0.6
Spain	60%	6.2	4.8
UK	60%	7.8	6.5
Poland	100%	11.7	7.2

Note: Poland, Portugal and France have substantial dispersed populations.

Major river catchments

Since the UWWTD requires phosphorus removal only for population centres above 10000 population equivalents, full implementation may not reduce inputs in sensitive catchments containing a large number of small towns or villages. In some parts of Europe this is not an issue, as even small population centres have been required to have phosphorus removal; Scandinavia is one example. In other parts of Europe it is not yet certain what the environmental authorities will require.

In order to investigate this issue, the population centres in some major catchments were identified, using a database provided by Harper Collins. In the database, each centre was associated with a population band, and this was used to derive the classification of [table 6.4](#). Reliable estimates of dispersed populations cannot be made from this data.

Table 6-4 Population in small centres for some major catchments

Catchment	Country	Number of population centres ¹			Estimated population		
		2k to 10k	10k to 100k	>100k	total millions	<2k dispersed ² %	2k to 10k %
Oder	Germany	59	41	2	3.5	4%?	8%
Seine	France	396	162	7	15.4	10%?	12%
Ebro	Spain	386	149	17	13.1	18%?	14%
Tagus	Spain	347	103	8	10.3	18%?	15%
Shannon	Ireland	102	42	1	3.2	18%?	16%
Po	Italy	678	354	16	22.4	4%?	16%
Rhine	Germany	1930	1020	49	60.9	4%?	16%
Elbe	Germany	1093	360	14	26.4	4%?	19%
Rhone	France	464	99	6	10.0	10%?	21%
Garonne	France	381	77	1	6.4	10%?	27%
Guadiana	Spain	360	86	4	6.4	18%?	28%
Loire	France	760	104	7	11.0	10%?	31%

Note 1. Derived from population centre data purchased from Harper Collins (Bartholemew's maps).

Note 2. From EEA, Environment in the EU at the turn of the century, chapter 3.5 water stress, fig.3.5.12. The figures applied to individual countries are uncertain.

The table shows that the median value of the population in centres between 2000 and 10000 is 16%, with three rivers (Garonne, Guadiana and Loire) having high proportions near to 30%.

7. LIFE CYCLE ANALYSIS

7.1 Introduction

This section presents a life cycle comparison between two options:

- a) Detergents have STPP as the builder,
- b) Detergents have zeolite A as the builder.

Comparisons are made for 2 sewage treatment options:

- i) Primary settlement and activated sludge, with denitrification to remove total nitrogen and chemical phosphorus removal. This represents a likely option for treatment of populations up to ~50000. A population of 20000 is assumed in the calculations. See figure 7.1
- ii) Primary settlement and activated sludge, with denitrification to remove total nitrogen and biological phosphorus removal. Chemical phosphorus removal can be used as a back up. This represents a likely option for treatment of populations above ~50000. A population of 200000 is assumed in the calculations. See figure 7.2.

A third option, of removing phosphorus as a sidestream in a form suitable for re-use, is also considered.

Three alternatives for sludge treatment and disposal are considered: use on agricultural land, incineration and landfill. All are important at present in at least some EU states.

The analysis takes account of:

- The energy and environmental impacts associated with the detergent builders

Information from existing LCA assessments is used selectively, along with some more recent information from a variety of sources.

- The energy and environmental impacts arising from the treatment process.

Each stage in the processes is modelled, to provide estimates of inputs and outputs of energy and materials.

This is not a full life cycle analysis as defined by the EEA framework (EEA 1998). However, conclusions can be drawn which are sufficient for the purposes of this report.

7.2 Processes for phosphorus removal from wastewater

A summary of the advantages and disadvantages of the two main process options is given in tables 7.2 to 7.4.

Table 7-1 Wastewater and sludge treatment processes used for the LCA comparison

Population equivalent	20000	200000
Wastewater treatment	Primary settlement Activated sludge with total N removal Chemical P removal ²	Primary settlement Activated sludge with total N & P removal Back up chemical P precipitation ^{1, 2}
Sludge treatment & disposal: option A	Thickening Transport off site to major sludge centre for eventual agricultural use	Thickening Digestion Dewatering Use on agricultural land as cake
Sludge treatment & disposal: option B	Thickening Transport off site to major sludge centre for incineration	Thickening Dewatering Transport off site Incineration at a larger sludge centre

Note 1. Chemical dosing to remove phosphorus will be assumed to back up biological removal, used only when necessary to achieve the UWWTD standard.

Note 2. It is assumed that chemicals are added just before entry to the aerobic part of the biological treatment process

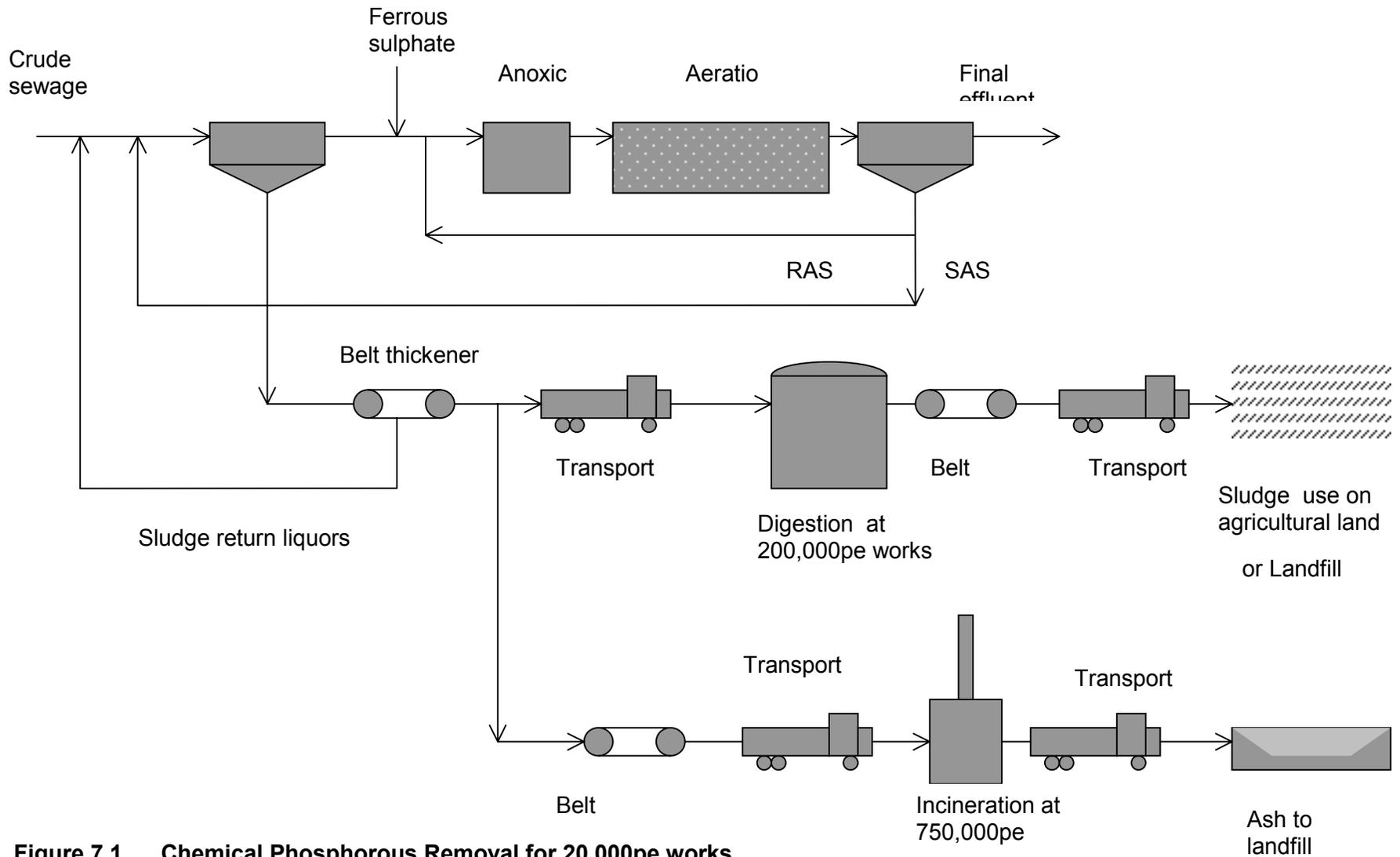


Figure 7.1 Chemical Phosphorous Removal for 20,000pe works

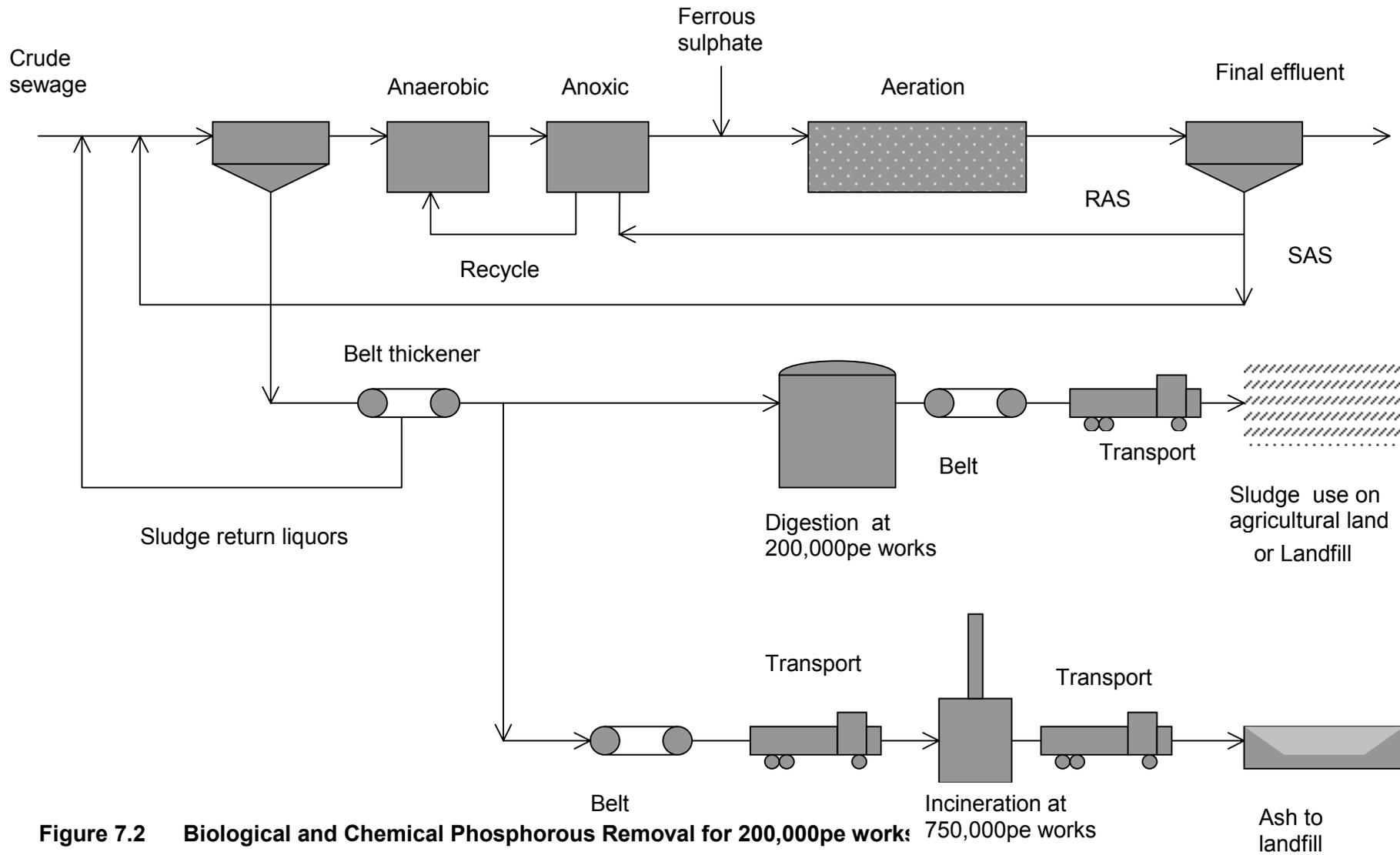


Figure 7.2 Biological and Chemical Phosphorous Removal for 200,000pe works:

Table 7-2 Comparison of the treatment processes

Option	Conventional chemical P removal	Biological P removal (with chemical back up)
P removal achievable	up to 95%	80% -95%
Average concentration P in treated effluent given conc. in crude sewage:		
15 mg/l	<1 mg/l	< 2 mg/l – may require chemical
11 mg/l	<1 mg/l	< 1 mg/l – can function without chemical
8 mg/l	<1 mg/l	< 1 mg/l – no chemical required
Sludge production quantity (g/pe/day) (Expect about 75 g/pe/d without P removal for primary + secondary).	20%-40% more than secondary activated sludge. Assume 90 g untreated; 63 g post-digestion	Similar to secondary activated sludge. Assume 75 g untreated; 50 g post-digestion.
P in sludge (% dry solids) (including primary sludge)	3% - 5% w:w dry solids	3% - 5% w:w dry solids
P availability as fertiliser from digested sludges: thermally dried are less; lime-treated sludges are more available	<1% - 30% dependent on measurement methods	15% - 100% dependent on measurement methods
Chemicals used	Precipitant, e.g. FeSO ₄ , FeCl ₃ Alum salts Metal :P molar ratio 1.5:1	Volatile Fatty Acid (VFA) may be needed as ready carbon source. (e.g. acetic acid)

Table 7-3 Pros and cons of chemical P removal

Advantages	Disadvantages
<ul style="list-style-type: none"> • Reliable well documented technique • Chemical costs can be reduced substantially if waste pickle liquors (ferrous chloride or ferrous sulphate) are available and can be used. • Controls are relatively simple using controls on metal dosing rates to maintain high P removal efficiency. • Installation at existing works is relatively straightforward. • Biological sludge can be processed in the same manner as in non-P removal systems. • Primary clarifier metal addition can reduce organic load to the secondary unit by 25%-35% by related removal of suspended solids and BOD. 	<ul style="list-style-type: none"> • Operating costs are higher than for biological systems. • More sludge is produced than by biological processes, which may overload existing sludge handling facilities. • Poorer dewatering qualities than sludges without added metals. • Tertiary filtration required to remove phosphorus in effluent suspended solids. • Effluents may become coloured if iron salts are used. • Potential for toxicity to the biological processes if the process is inefficient. • Phosphorus is not as available for agricultural use as comparable amounts of P fertilizers. • Increases metal loads on agricultural land.

Table 7-4 Pros and cons of biological P removal

Advantages	Disadvantages
<ul style="list-style-type: none"> • The amount of sludge generated is similar to conventional biological treatment processes. • Can be installed at existing 'plug-flow' activated sludge plants with minimal new equipment requirements, provided the plant has sufficient capacity. • Existing sludge handling methods can be used provided the risk of phosphorus resolubilisation are controlled. • No chemical additive costs, except for the 'PhoStrip' process, unless effluent polishing is included. • Some process arrangements combine nitrogen and P removal at virtually no additional costs. • Improved process control results in reduced risks of filamentous organisms. • The enhanced P content of sludge is as available for agricultural use as other similarly treated sludges. • Use of sludge for P source reduces soil P leaching. 	<ul style="list-style-type: none"> • P removal is limited by the BOD:P ratio of influent wastewater (except for PhoStrip system). In particular requires low molecular weight substrates available in the influent; suitable process design means this should not normally be an issue. • Secondary clarifier performance must be efficient to achieve effluent concentrations of 1 mg P/litre. • Cannot be readily used to retrofit fixed film processes (except for PhoStrip system). • Potential for P release during sludge handling, which would then be recycled to the head of the sewage treatment works. • Standby chemical treatment equipment may be required in case of low performance or failure of the biological system. • Sludge can have poor settling characteristics. • Increased retention time required in activated sludge plants. • Increased P in sludge enhances soil P index.

7.2.1 Results of the spreadsheet model

The model estimates the costs and energy used in wastewater and sludge treatment processes, for a range of influent sewage phosphorus concentrations (table 7.5). It is described in Appendix E, where some of the outputs are tabulated.

The model does not consider the full range of environmental and health impacts, such as those from greenhouse gases or toxic substances. Concerns over these impacts exist (e.g. emissions of dioxins from incinerators), but they are not directly relevant to the subject of this report.

Comparison between the process options shows that:

- Sludge incineration is both more expensive and has higher net primary energy (NPE) consumption than digestion and use on agricultural land. This is not surprising, and clearly there are other reasons why incineration is widely used.
- The annual cost and energy used are nearly independent of whether the detergents are built from STPP or Zeolite. Different assumptions on the value and availability of phosphorus in sludge, or the quantities of chemicals used, do not change this conclusion.

Table 7-5 Model outputs for process option 2A, sludge to agricultural land

% detergent built from STPP		0%	>~50%	100%
P concentration in crude sewage		8 mg/l	12 mg/l	15 mg/l
Annual cost ¹	euro/pe/year	6.11	6.25	6.36
NPE used ²	GJ/pe/year	0.11	0.11	0.11
Zeolite in crude sewage	t/year	161	69	0
P in crude sewage	t/year	184	276	345
P re-used ³	t/year	86	132	167
P lost	t/year	98	144	178

Note 1. Includes annual and investment costs.

Note 2. Net primary energy. A factor is used to convert energy used to NPE, depending on the form of the energy. Energy produced from digestion and the energy value of N & P in sludge are taken into account. Includes an allowance for the energy cost of construction.

Note 3. 60% availability of P assumed.

As further backing for this last point, consider the quantities of sludge produced in different situations (table 7.6). Statements have sometimes been made about the extra sludge that would arise from changing from STPP to Zeolite builders, or introducing phosphorus removal. Hard information is scarce, and calculations by WRc (Appendix F and table 7.6) suggest that the differences are not as great as has sometimes been claimed.

Table 7-6 Sludge production in biological sewage treatment

Household detergents	No P removal	Biological P removal	Chemical P removal
	g/pe/day	g/pe/day	g/pe/day
100% STPP based	86	87	98
50% STPP / 50% Zeolite	90		
100% Zeolite	93	89	99

The conclusion to be drawn is that any judgement of the relative merits of STPP and Zeolite as detergent builders should not rely on their effect on sewage or sludge treatment processes.

Sidestream phosphorus removal

Different processes have been proposed, some of which have been implemented on a limited scale. The Crystallactor process has been considered here as an example.

The activated sludge from final settlement is treated with acetic acid, to solubilise the phosphorus. After thickening, the liquor is treated with sodium hydroxide (or lime) and the phosphorus is precipitated. A high degree of phosphorus removal can be achieved. However at present the chemical costs are high. For a more detailed statement of the pros and cons, see below.

Advantages	Disadvantages
<ul style="list-style-type: none"> • Sludge production can be similar to non-P removal treatment. May significantly reduce sludge production (Krepro process). • P recovery eliminates or reduces potential for struvite formation in sludge treatment and pumping processes, reducing sludge operating costs. • Can be installed at existing works as additional process stream. • Provides a material that has a demonstrated economic value. • Reduces P load in sludge so improving the N:P ratio and reducing P overloading for agricultural recycling. • Reduces inert content in sludges so improving heat value and reducing slag for incineration. • Improves recycling efficiency of substance that has relatively limited economic availability; reduce environmental effects related to P extraction. • Low purity struvite or iron phosphate may be used directly or indirectly as fertilisers; apatite 	<ul style="list-style-type: none"> • Technology is new and varied; costs are high. • Most useful product has not been identified. • No clear destination for products yet. • Incomplete knowledge in use of P products directly as fertilizers. • Most processes require chemical supplementation; some require hazardous materials (sulphuric acid) or conditions (high temperature and pressures). • Additional to chemical or BPR process. • Can require sludge dewatering stage that may not otherwise be used.

Advantages	Disadvantages
<p>may be used as a high purity primary P source for conversion to higher value products.</p> <ul style="list-style-type: none"> • Low heavy metals content in recovered P compared to fertilisers. • May improve P removal efficiency by reducing P in sludge liquor return. • Improves the control of soil P when sludge is used as fertilizer. 	

7.3 Detergent builders – STPP

Past LCA studies include Morse et al (1994) and Landbank (1994). More recent information from ThermPhos (1999), one of the STPP manufacturers, has been used.

Information from the Landbank Report has not been used because:

- It is out of date, being based on the Whitehaven STPP production processes, which are no longer operating,
- It makes no allowance for the energy used in generating energy in a useful form. In particular energy is lost in generating electricity from fossil fuels (Fawer 1996 section 15). This means that Landbank's estimates of energy used are lower than they should be.

The ThermPhos STPP production process is shown in [table 7.8](#). It is one of 6 European manufacturers, and is not typical. The other manufacturers use the 'wet' process ([table 7.9](#)), similar to that assumed by Morse et al.

Morse et al's report, although 7 years old and based on information from 10 or more years ago, does consider energy correctly and provides a summary of the environmental impacts associated with phosphate and STPP production. The impacts include those of materials (e.g. sulphuric acid) used in the production processes, transport (in particular energy used) and waste streams. Morse et al's figures are shown in [table 7.7](#), alongside information provided by ThermPhos.

In order to update the information in Morse et al's report, the European Centre for the Study of Polyphosphates was approached with questions, in particular about the waste streams from STPP manufacture. In reply these questions, CEEP replied as follows (personal communication Chris Thornton, CEEP, to Rod Palfrey, WRc, 24/11/2001):

- *Detergent phosphates are responsible for <10% of gypsum production, since most phosphate produced is used in fertilisers, animal feeds and other industrial applications.*
- *Inert gypsum is either recycled into building materials, or landfilled to sites which can be landscaped and reforested after use. The heavy metals are in an immobile form. The radioactivity in the gypsum is sufficiently low to enable its authorisation as a building material (note 1).*

- *Heavy metals are removed from phosphate rock in two stages in the wet acid process. Around 90% will be transferred to the gypsum in an immobile form... The remaining metals are (except at one site) removed by purification processes in the case of detergent, food or industrial phosphates. These heavy metals are transferred to and stabilised in the gypsum stream (note 2).*

Note 1. Radioactivity. By product gypsum in the USA radiates on average 186 Bq/kg from uranium. For comparison radiation from uranium in clay bricks is stated to be 111 Bq/kg, and from granite rock 63 Bq/kg (USEPA 2000). WRc has not attempted a thorough investigation of this issue.

Note 2. Heavy metals can be released again in an aquatic environment, as has happened in the Baie de Seine. This should not be an issue at properly managed landfill sites.

In summary:

- The source of phosphate ore may be Russia, Morocco, Jordan or Florida. There are significant differences between ores in terms of their phosphate concentration, and the concentrations of other contaminants (such as Cd, U, F).
- For some ores, large quantities of 'tailings' are produced, i.e. waste material from concentrating the phosphate rock.
- Waste substances that require treatment and safe disposal include cadmium, radio-active uranium and fluoride. Currently they are immobilised in the waste gypsum and landfilled or used in construction materials.
- The environmental impacts from phosphate rock extraction and beneficiation, and in some cases from production of the crude green phosphoric acid, are outside the EU. Information on these, and environmental protection measures, is not easy to obtain.

Table 7-7 Impacts of STPP production

Parameter ¹	Morse et al ⁴	ThermPhos (1999)
Raw Materials Phosphate rock Sulphur (for H ₂ SO ₄) Limestone (for Ca(OH) ₂) Rock salt (for NaOH)	4.5 ² 0.75 1.0 1.4	2?
Water Process Cooling	>37.5 >125	
Key impurities in STPP Cadmium	0.2 mg /kg	
Energy	21 MJ / kg	14 MJ / kg for all phosphorus products. The Therm Phos process probably uses more energy. See note 3.
Key wastes Wastewater Phosphogypsum Other	>25 1.25 F, Cd, U ₃ O ₈ , Zn, NH ₃	Wastes to water, air and in solid form are quantified, with quantities in 1999 being generally less than in 1998, and significantly lower than in 1990. Targets for 2002 are shown. Process improvements have been made. The source of phosphate rock is an important factor that influences the quantities of hazardous materials.

Note 1. Units are kg / kg STPP, unless otherwise stated

Note 2. Highly dependent on where the rock is mined

Note 3. Not included in the 14 MJ/kg are (i) transport to the ThermPhos site and probably also (ii) the energy cost of chemicals not included and (iii) the energy may not be stated as net primary energy. The proximity of the ThermPhos plant to a nuclear power station complicates the argument.

Note 4. Morse et al's figures have been adjusted, as they assumed 0.8kg STPP and 0.2kg carbonates.

Table 7-8 STPP production – ThermPhos process

Process	Inputs	Output	Waste streams	Comments
<i>At point of extraction</i>				
Beneficiation	Phosphate rock Water	Concentrated phosphate rock	Tailings	The quantity of waste depends on the source. Said to be small for Moroccan rock, unknown for Russian rock
Transport	Fuel			
<i>At ThermPhos site</i>				
Conversion to phosphorus	Concentrated phosphate rock Coke Gravel Electrical energy	Phosphorus	Gas containing CO Phosphorus slag Iron slag	Gas can be burnt to produce energy Phosphorus and iron slags can also be used
Phosphoric acid production	Phosphorus Oxygen Water	Phosphoric acid		
STPP formation	Sodium hydroxide Heat	STPP	Offgas scrubber liquid	Heat is derived from CO derived from phosphorus production (as the element). Heat exchangers use the heat in the offgas

Table 7-9 STPP production – wet process

Process	Inputs	Output	Waste streams	Comments
<i>At point of extraction</i>				
Beneficiation	Phosphate rock Water	Concentrated phosphate rock	Tailings	The quantity of waste depends on the source. Said to be small for Moroccan rock, unknown for Russian rock
Transport	Fuel			
Conversion to phosphoric acid	Concentrated phosphate rock Sulphuric acid	Crude green phosphoric acid	Gypsum	Phosphogypsum contains metal impurities
Transport	Fuel			
Purification (1)	Crude green phosphoric acid Calcium hydroxide	Phosphoric acid with reduced sulphate	Gypsum	Gypsum used in mushroom fertiliser
Purification (2)	Phosphoric acid with reduced sulphate Concentrated NaOH	Sodium hydrogen phosphate solution	Precipitate/filtrate containing metals & fluoride	The waste stream is combined with other waste and forms an inert slag
Concentration & STPP formation	Sodium hydrogen phosphate solution Heat	STPP	Offgas scrubber liquid	Heat is derived from CO derived from phosphorus production (as the element). Heat exchangers use the heat in the offgas

7.4 **Detergent builders – Zeolite A**

Past LCA studies include Morse et al (1994), Landbank (1994) and Fawer (1996). Fawer et al (1998) summarise the results of Fawer (1996).

Fawer's report was commissioned by the ZeoDet sector group of the European Chemical Industry Council. Five European manufacturers of Zeolite A were represented, and the data were collected from these companies.

Information from the Landbank Report has not been used because it makes no allowance for the energy used in generating energy in a useful form. In particular energy is lost in generating electricity from fossil fuels (Fawer 1996 section 15). This means that Landbank's estimates of energy used are lower than they should be.

Morse et al's and Fawer's figures are compared (table 7.10). Both sets of figures are based on the processes summarised in table 7.11.

Table 7-10 Impacts of Zeolite A production

Parameter ¹	Morse et al ^{4,2}	Fawer ⁴	Other information
Raw Materials			
Bauxite	1	0.76	
Sand	1	0.47	
Rock salt	0.6	0.22	
Limestone	0.25	0.04	
Water			
Process	>13	Water for washing ore and cooling water not quantified.	Alcoa (2001) state that they now dewater red mud to 50% dry solids instead of 17%. Process water would therefore be less.
Cooling	>113		
Energy	24 MJ/kg	22 MJ/kg for slurry 26 MJ/kg for powder	
Key wastes	Red mud	Red mud (0.2 kg/kg, dry weight)	Toxic elements are not present.

Note 1. Units are kg / kg Zeolite otherwise stated

Note 2 Morse et al's figures have been adjusted, as they assumed 0.8kg Zeolite and 0.2 kg carbonates.

In summary:

- There is good agreement between two independent LCAs on the energy used in producing Zeolite A.
- The main waste stream is 'red mud' which is produced when the ore is digested. It is alkaline but does not contain toxic elements.

Table 7-11 Zeolite A production processes

Process	Inputs	Output	Waste streams	Comments
Transport ¹	Fuel			
Digestion and clarification to extract alumina	Bauxite Sodium hydroxide Heat	Aluminium rich (green) liquor	Red mud	
Precipitation	Aluminium rich (green) liquor	Aluminium hydroxide Al(OH) ₃ crystals	Spent liquor	Sodium hydroxide can be recovered from the spent liquor. For aluminium (but not Zeolite) production, the Al(OH) ₃ crystals are dehydrated.
Transport ¹	Fuel			
Solubilisation of aluminium hydroxide, crystallisation of Zeolite A	Aluminium hydroxide crystals Sodium hydroxide Sodium silicate Heat	Zeolite A crystals		

Note 1. The bauxite may be transported as ore and processed in Europe, or processed at the point of extraction and transported as aluminium hydroxide. Fawer (1996) assumes a mix of these options. Fawer assumes that the ore may come from Australia or Africa.

7.5 Detergent builders – Polycarboxylates

Polycarboxylates are derived from petroleum based feedstocks. A detailed LCA would be complex, and would not be appropriate here as PCAs are a comparatively minor component of Zeolite based detergents. Morse et al's work is assumed to apply ([table 7.12](#)).

Table 7-12 Impacts of polycarboxylate production

Parameter ¹	Morse et al ⁴	Comment
Raw Materials Oil	One of many products of refining	
Water Process Cooling	1 10	
Energy	5 MJ/kg	
Key wastes	Emissions to atmosphere associated with oil processing	i.e. Particulates, CO, CO ₂ , SO _x and volatile organic compounds

Note 1. Units are kg / kg PCA otherwise stated

7.6 Comparison between detergent builders

The results of the LCA are summarised in table 7.13 as a comparison between 1kg of STPP and 1kg of Zeolite A + 0.3kg of PCA.

Table 7-13 Comparison between STPP and Zeolite A

	STPP (1kg)	Zeolite A (1kg) / PCA (0.4kg)
Raw Materials	Phosphate rock (2 to 4.5kg) Sulphur (0.75kg) Limestone (1kg) Rock salt (1.4kg)	Bauxite (<1kg) Sand (<1kg) Limestone (<0.25kg) Rock salt (<0.6kg) Oil
Water – Process – Cooling	>37.5 litres >125 litres	>13.5 litres >117 litres
Energy	21 MJ	24 to 28 MJ
Wastes	Tailings where phosphate rock is mined. Phosphogypsum (1.25kg), containing Cd and U. These metals are in a chemically inert form.	Red mud (0.2kg dry weight, water content >50%). Emissions to atmosphere from oil processing.

8. CONCLUSIONS AND RECOMMENDATIONS

8.1 Overall Conclusions

A number of countries have been successful in reducing eutrophication through implementation of measures to reduce phosphorus loads. Notable examples are Lake Geneva in Switzerland, Lake Erie in the USA and Lake Endine in Italy. In all cases the results indicate that a phosphorus reduction of 70%-90%⁵ is necessary to significantly reduce eutrophication and improve trophic status.

A ban on the use of phosphate based detergents can achieve a phosphorus load reduction of up to 40% entering surface water bodies, which is not sufficient in isolation to result in any substantial improvements. Furthermore, improvements in wastewater treatment to fully comply with the UWWTD would only result in typical phosphorus reductions of around 30%. As demonstrated by Switzerland, the USA and Italy, the greatest improvements in lakes and rivers were observed where a combination of reduced detergent phosphorus and improved wastewater treatment were implemented, thereby achieving the required 70-90% reduction in external load.

The main sources of phosphorus entering surface waters are from municipal wastewater and agriculture. However, relative contributions vary depending on the nature of catchment landuse activities. For example, in areas without intensive agriculture (lake Geneva's catchment, lake Endine), municipal wastewater is the major source of phosphorus and in these areas improved wastewater treatment has been effective in reducing eutrophication. On the other hand, in catchments with intensive agriculture (e.g. lake Sempach in Switzerland, Wallonia, lower Rhine), agricultural inputs of phosphorus may represent a major source and a combination of measures including improved wastewater treatment and adoption of best land management practices should be employed.

Although the full implementation of the UWWTD will result in substantial reductions in phosphorus loads, discharges of wastewater without phosphorus removal would continue in sensitive areas, where the population is dispersed or in centres up to 10000 population equivalents. Further action to reduce phosphorus loads entering surface waters may be required in these areas.

Based on the results of life cycle analysis, Zeolite A was found to be a suitable alternative to STPP for use as a detergent builder. Only minor differences were observed in overall production cost in terms of energy used and sludge produced. Additionally, Zeolite A was found to be non toxic to aquatic fauna and humans and produces less toxic waste by-products when extracted from bauxite than phosphorus containing rocks (e.g. tailings produced include the heavy metals quantities are relatively minor. Furthermore, Zeolite A based detergents is generally accepted by EU Member States and consumers as an efficient and acceptable alternative to STPP based ones. The life cycle analysis concluded that 'any decision on the selection of a detergent builder should be based on other factors'.

⁵ Compared to 100% STPP based detergents and no nutrient removal from wastewater

The EU contributes to less than 10% of the world's STPP production, and employs approximately 1000 people. Therefore, while an EU wide ban on STPP use would direct STPP manufacturing to other large centres, such as China and India, the economic loss of this is not considered to be great in overall EU terms. Additionally, as the current EU capacity for Zeolite A production exceeds the actual production, it could be expected that increased production in this area would result in substantial employment and economic opportunities, with the only a small requirement for additional capital expenditure on infrastructure.

Excessive amounts of phosphorus has long been implicated in the eutrophication of surface water bodies. Therefore, to promote lake/river recovery and improve trophic status it is imperative that phosphorus loads entering surface waters are reduced. Based on the analysis of a number of countries, this phosphorus load reduction should be greater than 70% in order to achieve the above objectives. This can only be achieved through the implementation of a combination of limiting/banning the use of STPP based detergents and improving waste water treatment.

Zeolite A was shown to be a cost-effective alternative, both in terms of socio-economic and environmental impacts, to the use of STPP as a detergent builder in the EU. Therefore measures should be employed on an EU scale to restrict/ban the use of STPPs and switch to detergent builders based on Zeolite A.

8.2 Recommendations:

Based on the conclusions outlined above, the following recommendations are made:

- That a general ban on the use of STPP as a builder for household detergents be placed on all EU Member States;
- That EU Member States endeavour to reduce phosphorus loads entering surface waters in order to reverse the long term trend of eutrophication, through a combined approach of banning STPPs in household detergents and achieving full implementation of the UWWTD;
- That further investigations are undertaken on scattered populations and centres less than 10000 equivalents to determine the relative phosphorus contributions originating from these sources, after full implementation of the UWWTD, and what measures are needed and could be employed to reduce these contributions;
- That further investigations be undertaken within agricultural areas to identify 'best management practices', to reduce phosphorus loss to surface waters.

Table 8-1 Summary of river catchment case studies

Case	Starting point	Actions taken	Reduction of P inputs achieved	Effect on quality
Wallonia: Meuse & Schelt rivers	STPP based detergents Poor standard of sewage treatment	Change to Zeolite based detergents Improvements to sewage treatment begun	Not quantified	No reduction in eutrophication observed
France: Seine & Loire	STPP based detergents Sewage treatment does not remove P Intensive agriculture locally	Partial change to Zeolite based detergents Improvements to sewage treatment begun	~50% for the Seine Marginal for the Loire	No reduction in eutrophication observed
Germany: Rhine	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents Complete implementation of the UWWT directive including P removal	55-60%	See Netherlands, lake IJssel
Danube & Black Sea (Hungary)	Mainly STPP based detergents Poor standard of sewage collection & treatment	At an early stage	Not applicable	Unknown as yet.
Italy: Po river & N. Adriatic	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents Improvements to sewage treatment begun	30-40%	Partial improvement in quality of the N. Adriatic
Netherlands:	STPP based detergents Sewage treatment does not remove P Intensive agriculture	Change to Zeolite based detergents Sewage treatment removes P Measures to control agricultural P sources	50%	10% reduction in chlorophyll <u>a</u> in the IJsselmeer. This was less than expected, possibly due to reduced concentrations of toxic substances.

Table 8-2 Summary of lake case studies

Case	Starting point	Actions taken	Reduction of P inputs achieved	Effect on quality
France: Lac du Bourget	?	?	70%	Eutrophic to meso/eutrophic. Still in transition?
Germany: Lake Haussee	?	Ring sewer. No domestic sewage inputs	90%	Eventual recovery of the lake, >5 years after reducing P inputs
Italy: lago d'Iseo	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents P removal at main STW and diversion of some flow	60%	Lake still in transition from eutrophic condition
Italy: lago Endine	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents Ring sewer	80%	Lake still in transition from eutrophic to oligotrophic condition
Switzerland: lake Geneva	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents Sewage treatment removes P	60%	Significant improvement
USA: lake Erie	STPP based detergents Sewage treatment does not remove P	Change to Zeolite based detergents Major sewage treatment works remove P	85% from municipal wastewater, 50% overall	Significant improvement, recovery not complete

8.3 **policy options for controlling phosphorus**

Where a change from STPP based to Zeolite based detergents has been the only concerted action, reductions in quantities of phosphorus entering surface waters of 30-40% have been achieved and the beneficial effect on surface water quality has been small.

The question to be considered here is whether a change from STPP based to Zeolite A based detergents can be justified. The countries that would be effected by such a change are:

- EU member states: France, Greece, Portugal, Spain, UK.
- Accession states: Czech Republic, Hungary, Poland.

The case studies on France and Hungary have shown that (further) reductions in P inputs to surface waters are needed. These could and should be achieved through improved sewage collection and treatment. However, this is expensive, and the unit costs are particularly high for systems and plant that serve small communities. Implementing the UWWTD in the Accession States is expected to take a decade or more, and are at the limit of what can be afforded. Changing from STPP to Zeolite A as a detergent builder has no cost to consumers; clearly there is a cost to the STPP producers and a corresponding benefit to the Zeolite A producers.

The most economical way of achieving a given target for reducing P inputs is therefore *first* to change to Zeolite A as detergent builder, *while* improvements in sewage treatment are carried out.

APPENDIX A REFERENCES

Alcoa 2001	See the Alcoa web site www.alcoa.com.au/environment
Behrendt et al 1999	Nährstoffbilanzierung der Flußgebiete Deutschlands, Umweltbundesamt Forschungsbericht 296 25 515 UBA-FB 99-087
Bowker & Stensel 1990	
CEEP 2000	Étude du Phosphore, Proportion du Phosphore issu des Détergents dans les Eaux Continentales, Phase 1, Re-evaluation du Ratio Equivalent-Habitant en Phosphore, Geoplus, July 2000
CEFIC 2000	Zeolites for detergents as nature intended, ZEODET
CES 1991	Pollutants in Cleaning Agents. Report for UK Department of Environment by Consultants in Environmental Sciences Ltd.
CIPEL 2001	Rapports sur les études et recherches entreprises dans le bassin lemanique. Programme quinquennal 1996-2000. Campagne 2000.
Charlton et al 1999	Charlton M N et al, Lake Erie in transition: the 1990s, in 'State of Lake Erie – Past, Present & Future, pp.97-123, ed. M Munawar et al, Ecovision World Monograph Series, Backhuys 1999, ISBN 90-5782-018-8
Clark et al 2000	Clark G M et al, Water quality in the upper Snake River basin, Idaho and Wyoming, 1992-5. US Geological Survey monograph 2000-034982
Clement et al, 1998	Modelling the phosphorus retention of the Kis-Balaton upper reservoir A. Clement, L. Somlyódy and L. Koncsos** Water Science and Technology Vol 37 No 3 pp 113–120
Ecologic 2000	National Case Study on Policy Networks and the Implementation of the Urban Waste Water Treatment Directive 91/271/EEC in Germany, Case Study for TEP project, EU DG XII, January 2000
Edge 2001	Implications of Nutrient Removal from Sewage for Sludge Disposal Strategies, in AquaEnviro Seminar on Removal of Phosphorus and Recovery from Sludge, Leeds October 2001, ISBN 1-903958-02-4
EEA 1995	European Environment Agency 1995: Data reported by countries to the Topic Centre on Inland Waters for the report Europe's Environment - the second assessment.
EEA 1998	European Environment Agency 1998: Europe's Environment: The Second Assessment. Elsevier Science Ltd, Oxford, UK.
EEA 1998	Life Cycle Assessment. A guide to approaches, experiences and information sources. Updated 1998. Available on the EEA web site http://service.eea.eu.int
EEA 1999	European Environment Agency 1999: Nutrients in European Ecosystems. Environmental Assessment report No. 4. Office for the official publications of the European Communities, 1999.
EEA 2000	European Environment Agency 2000: Environmental Signals 2000. Environmental Assessment Report No. 6. Office for the official publications of the European Communities, 2000.
EEA 2000b	European Environment Agency 2000: Calculation of nutrient surpluses from agricultural sources. Technical Report 51.
EEA 2001	Europe's Environment: the Dobbris Assessment, May 2001
EEA undated	Environment in the European Union at the turn of the century, Chapter 3.5. Water Stress

EPDRB, undated	Environmental Programme for the Danube River Basin, Nutrient Balances for Danube Countries, project EU/AR/102A/91
Etienne, 2001	Sludge Production and Phosphorus Removal, Paul Etienne, Phosphorus Recovery Conference, Noordwijkerhout, Netherlands, 2001.
European Commission 2001	Name, Shame and Fame Seminar on City Sewage, February 2001
FAO 2001	FAO STAT home page http://apps.fao.org/
Farmer 1999	Implementation of the 1991 EU Urban Waste Water Treatment Directive and its Role in Reducing Phosphate Discharges, IEEC, London 1999
Fawer 1996	Life Cycle Inventory for the Production of Zeolite A for Detergents, EMPA Bericht Nr.234, 1996
Fawer et al 1998	Life Cycle Inventory for the Production of Zeolite A for Detergents, Fawer M, Postlethwaite D and Klüppel H-J, Int. J. LCA 3 (2) 71-74
Gachter R. and Meyer J. 1990	Mechanisms Controlling Fluxes of Nutrients Across the Sediment Water Interface in a Eutrophic Lake. In: <i>Sediments: Chemistry and toxicity of in-place pollutants</i> . Pp131-162.
Gachter R. and Wehrli B. 1998	Ten Years of Artificial Mixing and Oxygenation: No Effect on the Internal Phosphorus Loading of Two Eutrophic Lakes. <i>Environ. Sci. Technol.</i> , 32, pp 3659-3665.
Gaterall et al 2000	An Economic and Environmental Evaluation of the Opportunities for Substituting Phosphorus Recovered from Wastewater Treatment Works in Existing UK Fertiliser Markets, Gaterall M R, Gay R, Wilson R and Lester J N, <i>Environmental Technology</i> Vol. 21 pp.1067-1084, 2000
Gonsiorczyk T., Casper P. and Koschel R. 1997	Variations of Phosphorus Release from Sediments in Stratified Lakes. <i>Water, Air and Soil Pollution</i> . 99, pp427-434.
Hamm 1996	Wie und woher kommen die Nährstoffe in die Flüsse?, in Warnsignale aus Flüssen und Ästuaren – Wissenschaftliche Fakten, 105-110, Parey Buchverlag, Berlin.
Haskoning 1994	Environmental Programme for the Danube River basin: Danube integrated environmental study. Draft Report Phase 2, May 1994.
Hauthal H G 1996	Detergent Zeolites in an Ecobalance Spotlight, SOFW Journal Sonderdruck
Holmgren S. (1984)	Experimental lake fertilisation in the Kuokkel area, Northern Sweden. Biomass and algal composition in natural and fertilised subarctic lakes. <i>Internationale Revue der Gesamten Hydrobiologie</i> , 69, 781-817
Horne A.J. and Goldman C.R. 1994	<i>Limnology. Second Edition</i> . McGraw-Hill Publishers, USA.
IKSE 1995	Aktionsprogramm Elbe, International Kommission zum Schutz der Elbe, Magdeburg, 1995
Ireland EPA 2001	Urban Waste Water Discharges in Ireland – A Report for the Years 1998 & 1999, Republic of Ireland EPA (email info@epa.ie)
Joint Research Centre 2001	Criteria for the identification of freshwaters subject to eutrophication. Report prepared for DG Environment, January 2001. EI-JRC I-21020 Ispra Italy.
Kemezis, Paul 1999	No finale for phosphates. <i>Chemical Week</i> , v.161 no.4, January 27 1999, pp.31-32
Kohler 2001	Detergent phosphates and detergent ecotaxes: a policy assessment, prepared by Dr J Kohler of the University of Cambridge for CEEP
Kristensen 1998	Nutrient discharges from point sources in the European Union, NERI

	1998
Kroiss 1999	Water Protection Strategies – Critical Discussion in Regard to the Danube Basin, <i>Wat. Sci. Tech.</i> Vol.39 no.8 pp.185-192, 1999
Landbank 1994	The Phosphate Report
Magoarou 2000	European Commission Conference on Organic Waste Management, Stresa, Italy
Marsden M.W. 1989	Lake Restoration by Reducing External Phosphorus Loading: The Influence of Sediment Phosphorus Release. <i>Freshwater Biology</i> , Vol. 21, p139-162
Mawson S.J., Gibbons H.L. Jr., Funk W.H. and Hartz K.E. 1983	Phosphorus Flux Rates in Sediments. <i>Journal WPCF</i> . 55(8), pp1105-1110.
Monasterio 1996	Life Cycle Cost Analysis of Phosphorus Removal and Recovery from Sewage, Ortiz Monasterio, M Sc thesis, Imperial College London, 1998
Morse et al 1993	The Economic and Environmental Impact of Phosphorus Removal from Wastewater in the European Community, Morse G K, Lester J N and Perry R, CEEP 1993, ISBN 0 948411 08 2
Morse et al 1994	The Environmental and Economic Impact of Key Detergent Builder Systems in the European Union, Morse G K, Lester J N and Perry R, CEEP 1994, ISBN 0 948411 09 0
Morse et al 1995	The Life-Cycle Environmental Impact of Key Detergent Builder Systems in the EU, Morse G K, Perry R and Lester J N, the Science of the Total Environment Vol. 166 pp.179-192 1995
Mortimer C.H. 1941	The Exchange of Dissolved Substances Between Mud and Water in Lakes. <i>J. Ecol.</i> 29, pp280-239.
Mortimer C.H. 1942	The Exchange of Dissolved Substances Between Mud and Water in Lakes. <i>J. Ecol.</i> 30, pp147-201,
NERI 1997	Integrated Environmental Assessment on Eutrophication, NERI technical report no.207
Que Choisir 1999	Lessives, en poudre et pastilles; Que Choisir 363 Septembre 1999 pp48-51
RNDE 1998	L'Assainissement des Grandes Villes, obtainable from RNDE (the French Water Data Network) www.rnde.tm.fr
SENATOR 1996	Removal of Phosphate from Detergents in the Danube Basin, Final Report, project EU/AR/205/91
Sibbersen et al 1995	Phosphorus balance in European Agriculture – Status and policy Options. In SCOPE 54 - Phosphorus in the Global Environment. Ed. H. Tiessen.
Steen 1998	Phosphorus availability in the 21 st century. Management of a non-renewable resource. I Steen, <i>Phosphorus & Potassium</i> , 217, 1998
ThermPhos 1999	Environment and Safety Report 1999
Twinch A. J. 1986	The Phosphorus Status of Sediments in a Hypertrophic Impoundment (Hartbeespoort Dam): Implications for Eutrophication Management. <i>Hydrobiologia</i> 135, 23-34.
UBA Deutschland 2001	www.umweltbundesamt.org , Phosphoreinträge in Fließgewässer 1995
Vollenweider R.A. 1976	Critical Loading Levels for Phosphorus in Lake Eutrophication. Istituto Italiano Di Idrobiologia dott Marco De Marchie Memorie (Milan). 33, pp

	53-83.
Wetzel R.G. 1983	<i>Limnology. Second Edition.</i> Saunders College Publishers, USA.
Which? 1999	Coming Clean, Which?, p 24-27 September 1999, pub. UK Consumers Association.
Wilson et al 1994	The Phosphate Report, Wilson R and Jones B, Landbank Environmental Research and Consulting 1994, ISBN 0 9525639 0 8
Wilson et al 1995	The Swedish Phosphate Report, Wilson R and Jones B, Landbank Environmental Research and Consulting 1994, ISBN 0 9525639 0 8
Zessner et al 1998	Zessner M, Fenz R & Kroiss H, Wastewater Management in the Danube Basin, Water Science Technology, Vol.38 pp.41-49

APPENDIX B AGRICULTURAL AND INDUSTRIAL SOURCES OF PHOSPHORUS

Agriculture is a significant contributor to phosphorus pollution in many countries. This section presents information about the contribution that agriculture makes to phosphorus pollution in the EU countries, Hungary, Poland and the Czech Republic.

B.1 Land use in Europe

EEA (1999) reported that over 30% of Europe's land area is used for agricultural production. There are large variations in the amount of land used for agricultural purposes ranging from 85% in Denmark to 8% in Sweden. Within these figures there are also large variations in the amount of agricultural land that is used for arable farming. FAO (2001) land use data for each of the 15 Member States and 3 Accession countries are presented in [Table B.1](#).

Table B.1 Land use by country

Country	1998 Total Land Use Area	1989 Land Use (Agric.)	1998 Land Use (Agric.)	1989 Land Use (Arable)	1998 Land Use (Arable)	1989 % Arable of Agric.	1998 % Arable of Agric.
Austria	8.3	3.5	3.4	1.5	1.4	43	41
Belgium-Luxembourg	3.3	1.5	1.5	0.8	0.8	53	53
Denmark	4.2	2.8	2.7	2.6	2.4	93	89
Finland	30.5	2.6	2.3	2.5	2.2	96	95
France	55.0	29.5	29.9	17.9	18.4	61	61
Germany	34.9	17.6	17.4	12.0	11.9	68	68
Greece	12.9	8.1	9.1	2.9	2.8	36	31
Ireland	6.9	5.6	4.4	1.0	1.4	18	31
Italy	29.4	13.9	15.4	9.0	8.3	65	54
Netherlands	3.4	2.0	2.0	0.9	0.9	45	46
Portugal	9.2	3.7	3.6	2.9	1.9	78	53
Spain	49.9	25.8	30.1	15.6	14.3	60	47
Sweden	41.2	3.4	3.3	2.9	2.8	85	85
United Kingdom	24.2	17.9	17.5	6.7	6.3	37	36
Czech Republic	7.7	6.6	4.3	5.0	3.1	76	72
Hungary	9.2	6.3	6.2	5.1	4.8	81	78
Poland	30.4	18.5	18.4	14.4	14.0	78	76

Note: Source FAO 2001. Figures expressed as 1,000,000 ha.

Table B.1 shows that there are some very large differences in the proportion of arable land ranging from Denmark, Finland and Sweden (above 85% arable agricultural land) to Ireland and Greece (around 30%). Ireland is the only country where the proportion of arable land increased significantly between 1989 and 1998. There were quite large decreases in the proportion of arable land in Italy, Spain and Sweden between 1989 and 1998. There were small decreases in the proportion of arable land in the Hungary, Poland and the Czech Republic.

B.2 Fertiliser use and surpluses

Phosphorus fertiliser application levels provide another method of assessing possible impacts on aquatic environments. **Table B.2 and figure B.1** present phosphorus fertiliser consumption per unit area of agricultural land in EU countries, Hungary, Poland and the Czech Republic.

Table B.2 Phosphorus fertiliser consumption per unit area of agricultural land by country (FAO 2001)

Country	1991	1995	1999	1999
	000 tonnes			kg/ha/year
Austria	70	52	55	39
Belgium-Luxembourg	65	51	45	56
Denmark	76	49	41	17
Finland	84	73	53	24
France	1255	1031	966	53
Germany	519	402	420	35
Greece	176	136	119	43
Ireland	138	141	115	82
Italy	662	542	514	62
Netherlands	75	70	59	66
Portugal	76	71	77	41
Spain	502	510	643	45
Sweden	46	49	40	14
United Kingdom	365	390	320	51
Czech Republic		61	35	11
Hungary	24	56	40	8
Poland	151	302	297	21
EU	4110	3567	3467	46
EU and acc'n states		3986	3840	39

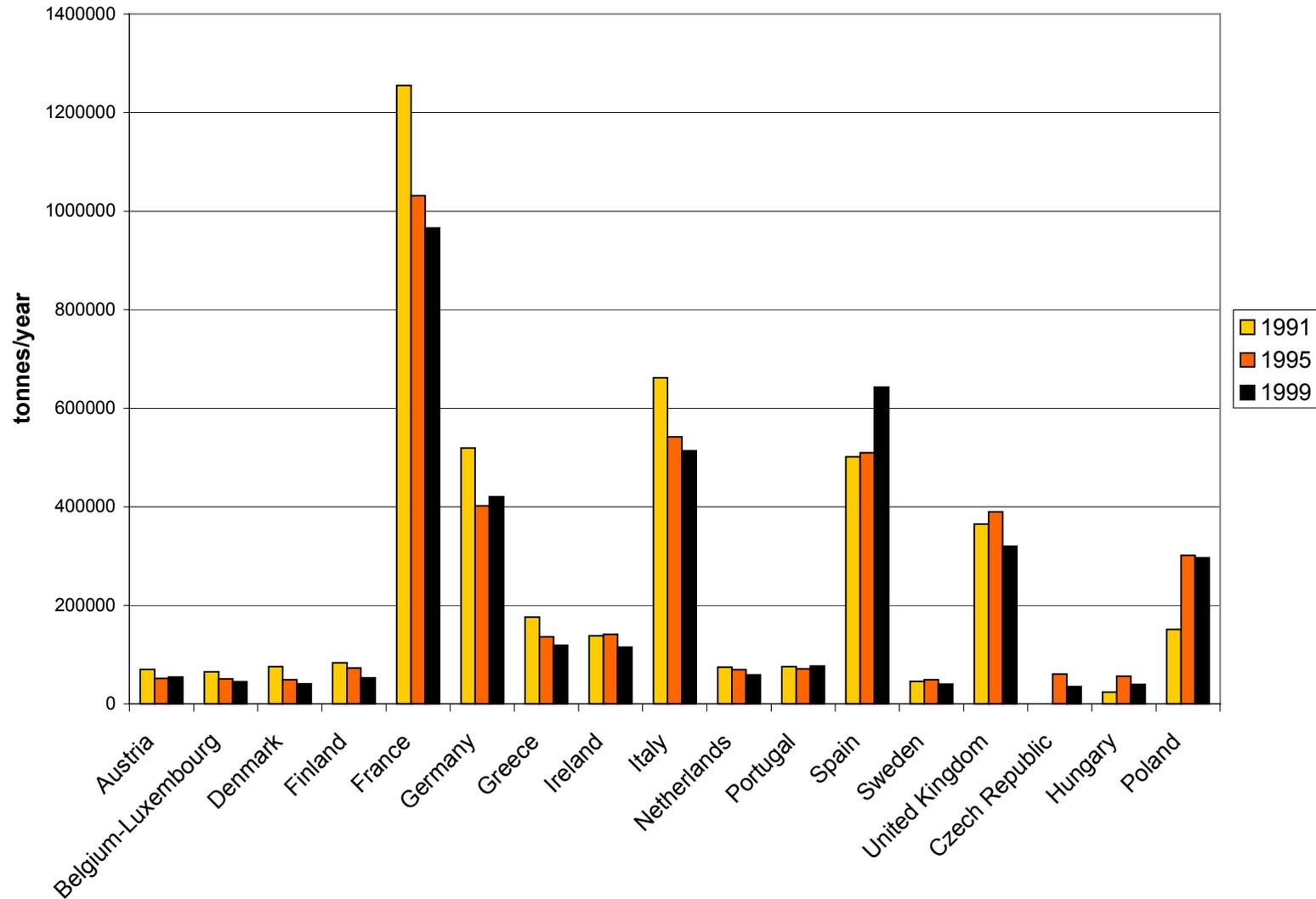


Figure B.1 Phosphorus fertiliser consumption in Europe

The data presented in [Table B.2](#) show a generally falling trend between 1991 and 1999. Notable exceptions are Hungary, Poland and Spain.

According to European Fertiliser Manufacturers Association figures phosphorus use by the agricultural sector will continue to decrease between now and 2010. Predictions are that by 2010 phosphorus use by EU countries will be 50% lower compared to peak consumption figures in the 1970's and 1980's. The largest decreases are expected to occur in the Netherlands and France.

Even with the reducing application of phosphorus, there is an annual surplus of phosphorus which is the difference between the total quantity put in, and the outgoing quantity in agricultural products. In the long term phosphorus surpluses can result in harmful losses to water (EEA 2000b). Generally the greater the surplus the greater the loss of nutrients to surface water. In the UK the average P surplus over the total agricultural area is about 10 Kg/ha/yr. The greatest risk of a localised phosphorus surplus occurs in arable soils receiving manure from intensive pig and poultry units (MAFF 2001 unpublished data).

Compounds of phosphorus are seldom washed out from soils. Phosphorus enters waterways mainly through soil erosion (62% of diffuse sources in Germany) and through surface runoff (24%) (UBA Deutschland 2001).

B.3 Livestock Numbers in EEA member countries

Across Europe livestock numbers have been fairly stable since 1990, with a slight decrease in cattle numbers and an increase in chicken numbers. There have been major trends in individual countries (figures [B.2 to B.5](#)). Spain is the exception, where livestock numbers of all 4 types have increased.

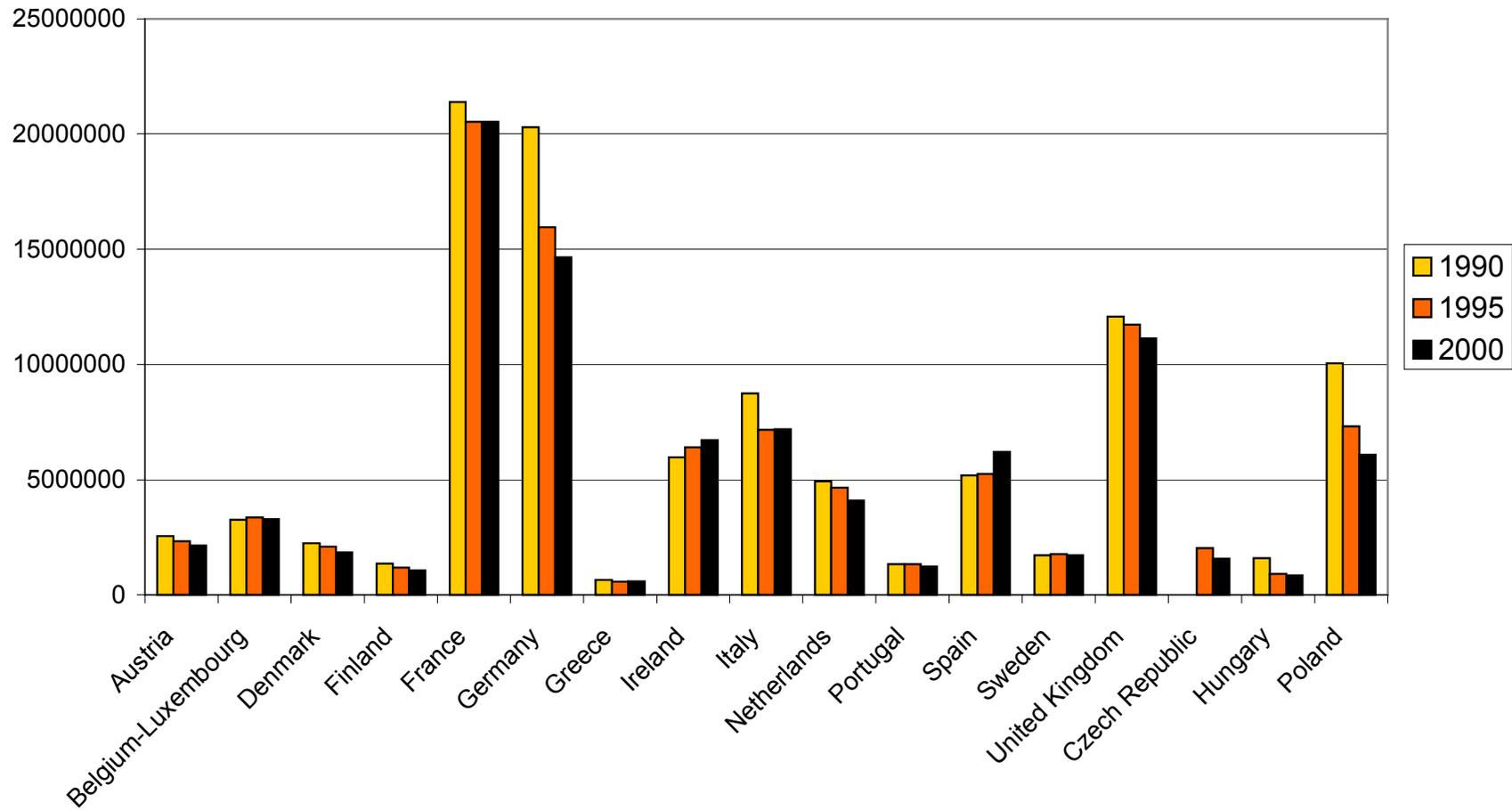


Figure B.2 Cattle numbers, 1990-2000 – EU and accession states

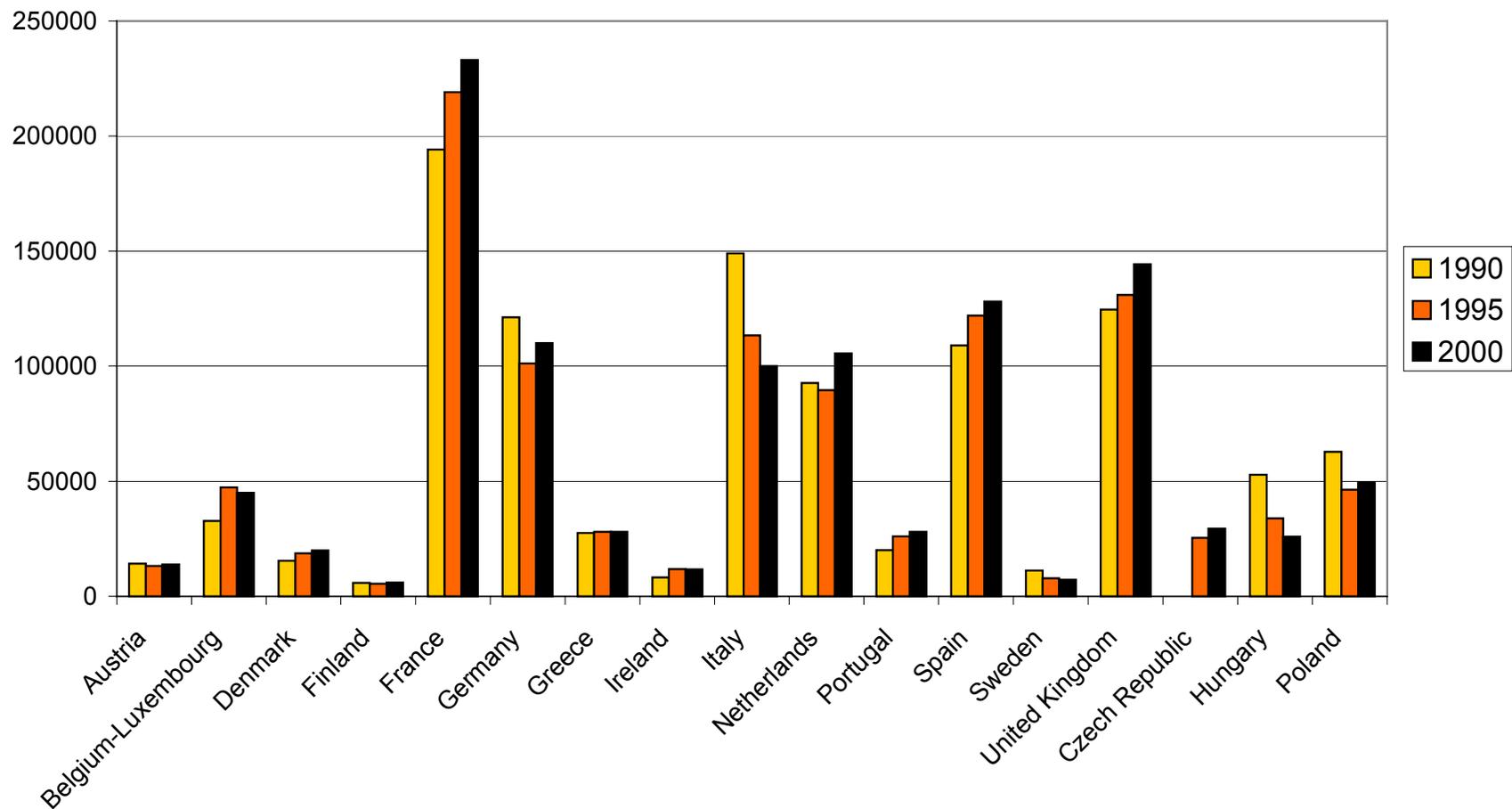


Figure B.3 Chicken numbers (000s) 1990-2000 – EU and accession states

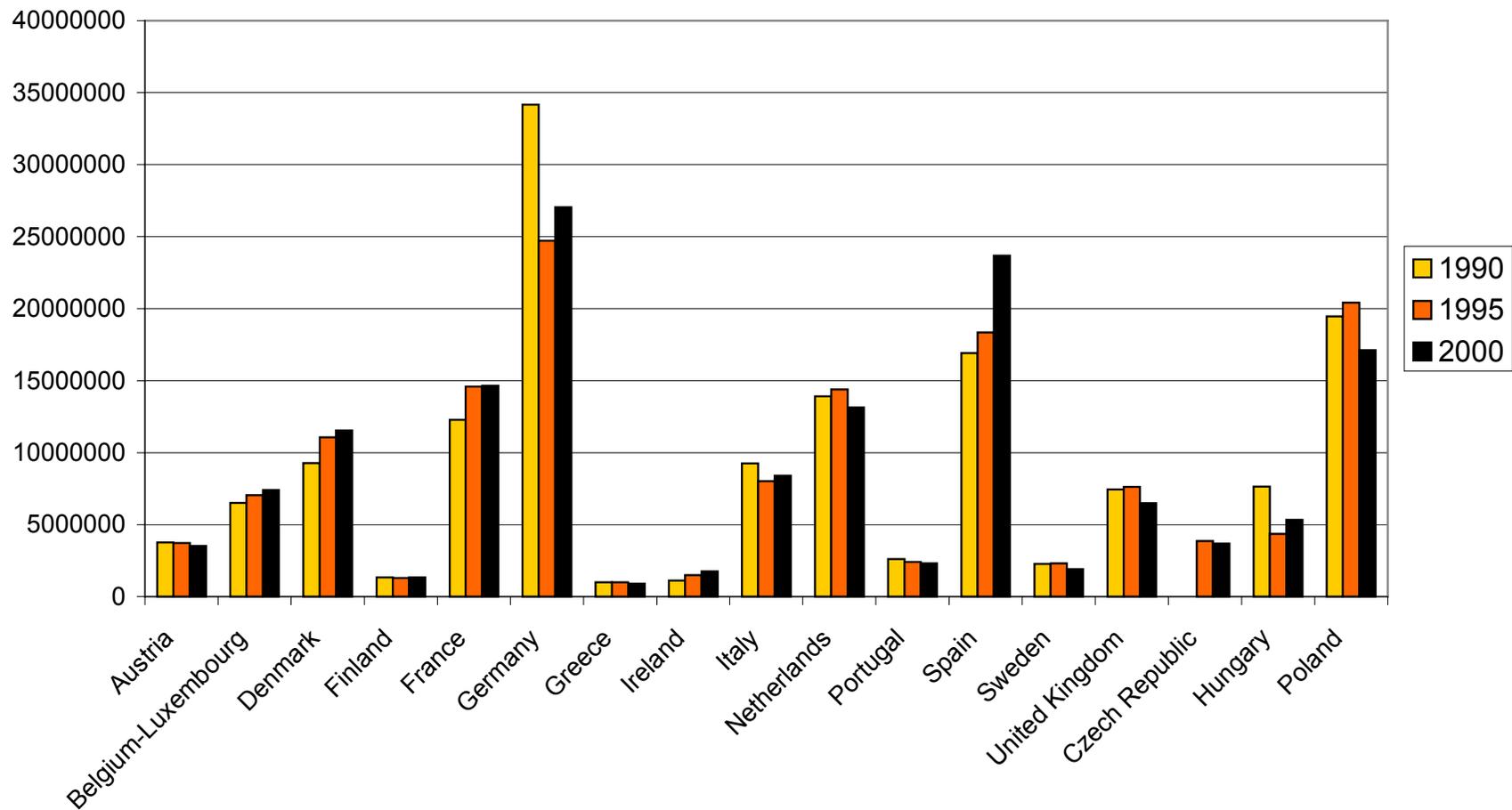


Figure B.4 Pig numbers 1990-2000 – EU and accession states

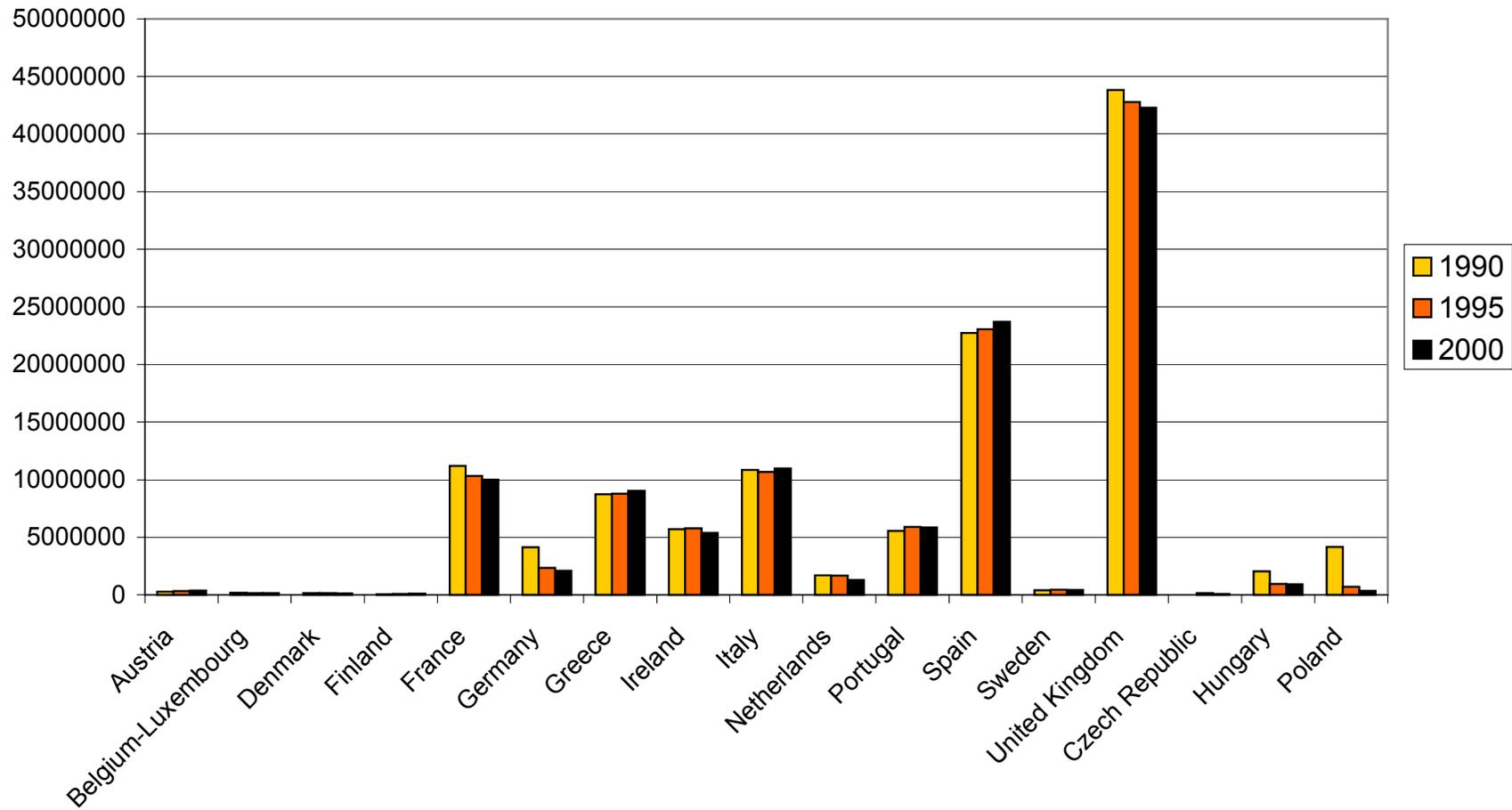


Figure B.5 Sheep numbers 1990-2000 – EU and accession states

B.3 Phosphorous balance in European agriculture

Animals represent one of three compartments in the agricultural phosphorus cycle, the others being soils and crops. Sibbersen and Metzger (1995) used a simple model to describe phosphorus flows in agriculture at a national scale. Included in this model is phosphorus loss to soil through excreta. **Table B.2** shows their estimate of the amount of phosphorus excreted by cattle, pigs and poultry in each country.

The estimated phosphorus loss varies considerably between species and between individuals within species (Sibbersen and Metzger 1995). This is due to differences in farming practice, including:

- stock numbers per hectare,
- whether ruminant animals (cattle and sheep) are fed on purchased feed or on grass,
- whether non-ruminant animals (pigs and chickens) are in factory farms.

Sibbersen et al note that the stocking rate (i.e. number of animals per hectare) is higher in Europe than in any other region of the world, and that within Europe it is highest in the Netherlands, followed by Belgium. They also note that:

- 'Phosphorus in mineral fertilisers is ideally used as a supplement to P in animal manure. One should therefore expect little use of mineral phosphate fertilisers in countries producing much excreta P... The opposite, however, is observed...'
- Using data from 1989, there was a net addition of phosphorus to soil in every European country, with an average of 13.7 kgP/ha/year. There were high values (>~50) in the Netherlands and Belgium, and low values (~7) in Spain and Portugal.
- '[Surplus] P, with its lower mobility, largely accumulates in the surface soil. This accumulation is beneficial if the P fertility of the land needs improvement. However, in parts of Europe the soil P content is now much greater than that needed by the crops. This increases the risk for P losses to the hydrosphere by leaching and erosion. In large areas of the Netherlands the soil may now be saturated with P so that P added in excess of crop uptake leaches from the soil...'

Table B.2 Phosphorus excreted by cattle, pigs and poultry in each country

Country	Phosphorus loss to ground in excreta (kg P/ha/yr)		
	Cattle & sheep	Pigs	Poultry
Austria	7.0	3.0	0.4
Belgium & Luxembourg	22.4	13.1	2.1
Denmark	10.1	10.8	0.7
Finland	6.1	1.8	0.3
France	6.9	1.5	0.7
Germany	12.5	6.4	0.8
Greece	2.0	0.5	0.3
Ireland	8.3	0.6	0.2
Italy	6.6	2.2	1.2
Netherlands	31.7	25.1	4.8
Portugal	3.7	1.5	0.5
Spain	2.1	1.6	0.5
Sweden	5.7	2.3	0.4
UK	6.8	1.4	0.9
Czech Republic	7.3	3.6	0.7
Hungary	3.1	4.4	1.0
Poland	5.3	2.5	0.4

Note: Adapted from Sibbersen and Metzger (1995).

B.4 Quantities of phosphorus in surface water from agriculture

Overall conclusions

Phosphorus transfer from soils to water is a potentially important factor influencing nutrient levels and eutrophication. Recently studies have shown that

- there is a continuing build up of phosphorus in soils throughout Europe,
- significant quantities of phosphorus can enter surface waters from erosion, and from leaching particularly in areas with high fertiliser inputs that are saturated with phosphorus e.g. Brittany.

Estimating phosphorus erosion and leaching rates is complicated because the process is influenced by a range of factors including soil and land use type, hydro-geological and climatic conditions. Loss rates from agricultural land in northern temperate Europe are estimated to be between 0.2 and 1.8 kg P/ ha/ year, with the majority between 0.3 and 1.0 (EEA 2001 chapter 14). Loss rates from catchments with natural vegetation (mostly forest) are between 0.05 and 0.1 kg P/ ha/year.

However, even if the current downward trend in phosphorus fertiliser use continues, it seems likely that there will be continue to be a net addition of phosphorus to soil. The quantity of

phosphorus entering surface waters is therefore unlikely to decrease in the foreseeable future, and may well increase.

Specific cases

Several case studies of particular areas or catchments have been done. Some of these are summarised in table B.3. Inputs from agriculture, and other non-point sources, generally have to be estimated, and are subject to considerable uncertainty.

- Domestic wastewater and agriculture are both major sources of phosphorus. Their relative importance varies with location.
- For the Rhine, the total input from wastewater treatment plants was reduced by 50% between 1985 and 1995, through the introduction of nutrient removal.
- Inputs from scattered dwellings and sewer overflows are probably minor where there are major cities whose wastewater treatment is without nutrient removal. They may be important sources where wastewater treatment removes phosphorus, as at Odense (or where a high proportion of dwellings are not connected to sewer).

Table B.3 Phosphorus inputs – specific cases

Catchment or area	Data source	Year	Description of catchment/area	Proportion of phosphorus by source			
				Agriculture	Industry	Domestic wastewater	Atmosphere & background
Rhine	NERI 1997	1995	Downstream of Bodensee	32%	16%	46% ¹	7%
Germany	UBA	1995	High degree of nutrient removal from urban wastewater	49%	10%	29% ¹	2%
Odense Fjord, Denmark	NERI 1997	1995	All wastewater treatment has nutrient removal	18%	1%	<ul style="list-style-type: none"> • Treated wastewater 22% • Scattered dwellings 21% • Sewer overflows 13% 	23%
Stratford Avon	NERI 1997	1995	Secondary wastewater treatment, not nutrient removal	30%	Minor	65% ¹	6%
Mayenne	CEEP 2000	?		41%	42%	17%	-
Upper Rhone	CEEP 2000	?		15%	46%	39%	-
Charante	CEEP 2000	?		64%	17%	18%	-
Denmark	EEA 2001, chapter 14	?	Densely populated. Wastewater treatment includes P removal.	17%	small	<ul style="list-style-type: none"> • Treated wastewater 50% • Scattered dwellings 17% 	17%
Sweden	EEA 2001, chapter 14	?	Sparsely populated. Wastewater treatment includes P removal.	<10%	small	25-30%	65%
River Po	EEA 2001, chapter 14	?	Densely populated	30%	<5%	>~65%	small

Note 1. Including industries that discharge to sewer.

APPENDIX C PHOSPHORUS DISCHARGES TO SURFACE WATER FROM MUNICIPAL WASTEWATER

Table C.1 Municipal Wastewater Treatment – Current Situation

Country	Total population (millions)	% of population							
		Not connected	Untreated	Primary	Secondary	Nutrient removal			
						Any	N only	P only	N & P
Austria	7.9	25%	0%	0%	27%	48%	0%	2%	47%
Belgium	10.1	0%	10%	0%	49%	41%	0%	0%	41%
Denmark	5.1	10%	0%	0%	2%	88%	0%	0%	88%
Finland	5.1	22%	0%	0%	1%	77%	0%	77%	0%
France	57.0	49%	4%	10%	16%	21%	16%	0%	6%
Germany	81.6	7%	2%	6%	6%	80%	0%	0%	80%
Greece	10.5	42%	9%	40%	9%	1%	0%	0%	1%
Italy	56.8	18%	11%	2%	36%	31%	0%	0%	31%
Luxembourg	0.4	1%	10%	5%	76%	8%			
Netherlands	15.4	3%	0%	1%	23%	73%	0%	60%	13%
Portugal	10.8	43%	39%	5%	12%	1%	1%	0%	0%
Republic of Ireland	3.5	31%	24%	26%	14%	4%	0%	0%	4%
Spain	38.5	18%	8%	20%	41%	13%	3%	3%	7%
Sweden	8.8	13%	0%	0%	0%	87%	0%	48%	39%
United Kingdom	57.5	5%	8%	8%	61%	20%	0%	18%	2%
Czech Republic	10.3	25%	4%	0%	59%	11%	8%	0%	4%
Hungary	10.4	40%	21%	7%	31%	1%	0%	1%	0%
Poland	38.7	35%	9%	10%	28%	18%	0%	18%	0%

Data sources: Country fiches from EU Name, Shame and Fame presentation, 2001, and data presented by individual countries to the EU. Also 'Urban Wastewater Discharges in Ireland, 1998-99, Irish EPA.

Table C.2 Population by size of centre

Country	Total pop.		Percentages		
	000s	dispersed	<10k	10k to 100k	>100k
Austria	8080	18.7%	35.0%	19.0%	27.2%
Belgium	10240	0.0%	12.6%	65.6%	21.9%
Denmark	5330	33.2%	18.4%	26.4%	22.0%
Finland	5180	19.3%	20.0%	32.7%	28.0%
France	58740	30.8%	21.6%	30.0%	17.7%
Germany	82690	0.2%	18.2%	41.1%	40.5%
Greece	10600	19.1%	11.9%	25.0%	44.0%
Italy	57460	10.3%	18.6%	41.8%	29.3%
Luxembourg	436	21.0%	36.6%	42.4%	0.0%
Netherlands	15870	0.3%	18.1%	48.5%	33.1%
Portugal	9790	44.5%	12.3%	22.2%	20.9%
Republic of Ireland	3710	44.8%	10.8%	13.2%	31.2%
Spain	39500	7.6%	17.8%	34.0%	40.6%
Sweden	8870	20.1%	18.1%	36.7%	25.1%
United Kingdom	59730	6.4%	14.6%	41.1%	37.9%
Czech Republic	10280	20.8%	20.8%	36.0%	22.4%
Hungary	10040	7.0%	28.3%	34.7%	30.0%
Poland	38640	33.9%	5.3%	28.9%	31.9%

Source. List of population centres from Harper Collins (Bartholomew maps). Exact populations have been used for major cities, and the mid points of population bands for minor towns.

Table C.3 Assumed P discharged for different treatment types

Treatment type	P discharged	
	% of inflow	mg/l
Dispersed population	50%	
Wastewater collected but not treated	100%	
Primary treatment	80%	
Secondary treatment	60%	
P removal - 10k to 100k population		1.5
P removal - >100k population		0.8

Table C.4 Future, UWWTD compliant, wastewater treatment

Country	% of population in sensitive area	Degree of treatment					Nutrient removal
		Dispersed	Untreated	Primary	Secondary		
Austria	100%	19%	0%	0%	7%	75%	
Belgium ^a	100%	0%	0%	0%	13%/0%	87%/100%	
Denmark	100%	10%	0%	0%	2%	88%	
Finland	100%	22%	0%	0%	1%	77%	
France ^a	705	31%	0%	0%	21%/36%	48%/33%	
Germany	100%	7%	0%	0%	14%	80%	
Greece ^a	70%	19%	0%	0%	24%/33%	57%/48%	
Italy ^a	60%	10%	0%	0%	36%/47%	54%/43%	
Luxembourg	100%	21%	0%	0%	37%	42%	
Netherlands	100%	3%	0%	0%	5%	92%	
Portugal ^a	20%	45%	0%	0%	44%/47%	11%/9%	
Republic of Ireland ^a	60%	31%	0%	0%	35%/42%	33%/27%	
Spain ^a	60%	8%	0%	0%	37%/48%	55%/45%	
Sweden	100%	13%	0%	0%	0%	87%	
United Kingdom ^a	60%	5%	0%	0%	39%/48%	56%/47%	
Czech Republic	100%	21%	0%	0%	21%	58%	
Hungary	100%	7%	0%	0%	28%	65%	
Poland	100%	34%	0%	0%	5%	61%	

Nutrient removal includes both N and P removal.

Note a. The two estimates for 'UWWTD fully implemented' represent different assumptions about treatment standard for population centres up to 10000 in sensitive areas: secondary treatment only / nutrient removal.

Table C.5 Phosphorus discharges under different scenarios

Country 000 tonnes/year	Current wastewater treatment		UWWTD fully implemented		Comment WWT	Current detergent content	Assumed detergent quantity
	Current detergent builders	Only Zeolite builders	Current detergent builders	Only Zeolite builders			
Austria	1.9	1.9	1.2	1.2	High standard	100% Zeolite	
Belgium ^a	3.0	3.0	1.3	1.3		100% Zeolite	
Denmark	0.7	0.6	0.7	0.6	High standard	20% STPP	
Finland	0.8	0.7	0.8	0.7	High standard	10% STPP	
France ^a	24.3	17.7	21.6	15.7		50% STPP	Less than now
Germany	13.3	13.3	12.2	12.2	High standard	100% Zeolite	
Greece ^a	6.2	4.5	3.4	2.5		50% STPP	
Italy ^a	17.6	17.6	14.6	14.6		100% Zeolite	
Luxembourg							
Netherlands	2.7	2.7	1.8	1.8	High standard	100% Zeolite	
Portugal ^a	6.9	4.5	4.9	3.2		70% STPP	More than now
Republic of Ireland ^a	1.7	1.7	1.1	1.1		100% Zeolite	Less than now
Spain ^a	21.7	15.0	14.3	9.9		60% STPP	
Sweden	1.2	1.1	1.2	1.1	High standard	15% STPP	
United Kingdom ^a	28.3	21.1	19.4	14.5		45% STPP	Less than now
Czech Republic	5.2	3.5	3.1	2.1		65% STPP	More than now
Hungary	6.4	4.2	2.9	1.9		70% STPP	More than now
Poland	21.9	13.4	11.7	7.2		85% STPP	Less than now

Assumed future detergent use: 8 kg/person/year detergent throughout the EU and Accession States.

Table C.6 Phosphorus discharges to sensitive areas – selected countries

Country	Assumed % population in sensitive areas	Population	Population in sensitive areas					UWWTD Option 1: 2ary treatment pe<10k		UWWTD Option 2: P removal pe<10k	
			Total	Sensitive	Dispersed	2k to 10k	10k to 100k	>100k	Current detergent builders	100% zeolite	Current detergent builders
		millions	millions	millions	millions	millions	millions	000 tonnes/year	000 tonnes/year	000 tonnes/year	000 tonnes/year
France	70%	57	39.9	12.3	8.6	12.0	7.0	14.9	11.4	8.4	6.9
Portugal	20%	10.5	7.35	1.4	0.9	1.8	3.2	0.8	0.6	0.6	0.4
Spain	60%	38.5	23.1	1.7	4.1	7.8	9.4	6.2	4.8	2.9	2.6
UK	60%	57.5	34.5	2.2	5.0	14.2	13.1	7.8	6.5	4.1	3.9
Poland	100%	38.7	38.7	13.1	2.0	11.2	12.3	11.7	7.2	10.7	6.6

APPENDIX D USA NATIONAL WATER QUALITY INVENTORY

National Water Quality Inventory

The NWQI provides an overview of current water quality conditions relative to *the “beneficial uses”* that the States and jurisdictions have established for their water resources, as required under the CWA. (See Annex for details of beneficial uses)

The National Water Quality Inventory first conducted in 1972, and now conducted biannually to be collated by the EPA for presentation to Congress. Using these inventories the progression of nutrient pollution can be traced in the 1990’s

In 1988 the definition of nutrient pollution was expanded to reporting on nutrient impairment in lakes, streams and estuaries. Impairment is measured as the inability to support the traditional uses of the water.

The definition of nutrients includes nitrates found in sewage and fertilisers and phosphates found in detergents and fertilisers.

Using the criteria below the water body is assessed using a combination of data. Monitored assessments are based on recent monitoring data collected during the past 5 years. Evaluated assessments are based on qualitative information or monitored information more than 5 years old. Numeric water quality criteria establish the minimum physical, chemical, and biological parameters required to support a beneficial use.

Criteria	Definitions
Fully Supporting All Uses	Good Water quality meets designated use criteria.
Threatened for One or More Uses	Good Water quality supports beneficial uses now but may not in the future unless action is taken
Impaired for One or More Uses	Impaired Water quality fails to meet designated use criteria at times
Not Attainable	The State, Tribe, or other jurisdiction has performed a use-attainability analysis and demonstrated that use support is not attainable due to one of six biological, chemical, physical, or economic/social conditions specified in the <i>Code of Federal Regulations</i> .

RIVERS

1992

Of the total assessed river miles (642 881) 48% (308 583) were found to be impaired to some degree with agriculture the highest single polluter, accounting for 160 000 miles of rivers while municipal point sources account for only 15% of impaired river miles.

The total of river miles assessed in each inventory is considerable when it is realised that only 1.3 million river and stream miles are perennial waters that flow year round. The remaining 2.3 million miles or so are intermittent or ephemeral, and are dry for some or most of the year.

1994

Of the total 3.5 million miles of river, 615 806 miles or 17% were surveyed. Of the surveyed river miles 224 236 miles were found to be impaired.

In 1994 nutrients impaired approximately 51 574 (23%) of all impaired river miles (224 236 miles), and were ranked third lead pollutant. This compared to the two highest contributors siltation and bacteria which each accounted for 35% of impaired river.

1996

Of the 3.6 Million miles of rivers and streams, 693 905 river miles were surveyed, with agriculture reported as the leading source of impairment in rivers, contributing to impairment of 25% of the surveyed river miles.

1998

Of the total 3 662 255 miles of river in the United States, the State authorities assessed 23% or 840 402 miles of rivers and streams. It was found that 291 263 miles of river were impaired. For the subset of assessed waters, 65% were rated as good and 35% as impaired.

Nutrient pollution accounted for 84 071 miles of impaired river, or 29% of the total length of river impaired. The most likely source of phosphorus originating from detergents, Municipal Point Sources, accounts for 29 087 (35% of nutrient impairment) miles of impaired river, but only 4% of total river impairment. The largest contributor to the impairment of rivers is agriculture, which accounts for 60% of impaired river.

Agriculture is listed as a source of pollution for 170 750 river and stream miles, or about 20% of assessed river and stream miles.

LAKES

1992

Of the total 40 million lake acres 46% or 18 million acres were assessed. Of this 46% nutrients were the second highest polluter after metals, with 40% of assessed lake acres being impaired by nutrients. The highest source of pollution in lakes was reported as agriculture accounting for 56% or 3 million lake acres of pollution.

The trophic state of lakes is also reported and in 1992 32% of assessed lakes were found to be eutrophic. Forty-one States also assessed trophic status in 11 477 of their lakes. These States reported that 17% of the lakes they assessed for trophic status were oligotrophic, 35% were mesotrophic, 32% were eutrophic, 7.5% were hypereutrophic, and 8.5% were dystrophic.

1994

States reported that 18% of the lakes they surveyed for trophic status were oligotrophic, 32% were mesotrophic, 36% were eutrophic, 6% were hypereutrophic, and 3% were dystrophic.

This information may not be representative of national lake conditions because States often assess lakes in response to a problem or public complaint or because of their easy accessibility. It is likely that more remote lakes which are probably less impaired are underrepresented in these assessments.

Of the total 40.8 million acres 17.1 million acres (42%) were surveyed and 17% were found to be impaired, of this figure nutrients were responsible for 26 847 square miles (43%). It was further reported that extra nutrients pollute 2.8 million lake acres (which equals 43% of the impaired lake acres).

Agriculture was by far the biggest source of pollution accounting for 50%, followed by Municipal Point Sources with 19%.

1996

States reported that 16% of the lakes they surveyed for trophic status were oligotrophic, 38% were mesotrophic, 36% were eutrophic, 9% were hypereutrophic, and less than 1% were dystrophic.

Nutrients was the joint highest ranking course of pollution (along with metals) with 20% of assessed lake acres being impaired by nutrients

The National Water Quality Inventory of 1996 reported that of the lakes inventoried 36% were eutrophic.

1998

In 1998, 32 states reported that 17% of the 7 373 lakes they assessed for trophic status were oligotrophic, 33% were mesotrophic, 38% were eutrophic, 12% were hypereutrophic, and less than 1% were dystrophic.

Of the total lakes, 41 593 748 acres, 17 390 370 acres (42%) were assessed with 7.9 million acres being impaired. It is reported that excess nutrients pollute 3.5 million lake acres which equals 20% of the assessed lake acres and 44% of the impaired lake acres.

Municipal Point Sources of pollution accounted for 5% of the pollution in all lakes assessed and 11% of impaired lake acres.

ESTUARIES

1992

In 1992 roughly 75% of the USA's estuarine waters with 32% being reported as impaired, of which 55% of the total assessed (8 572 square miles) were reported as being impaired by nutrients. The highest-ranking pollutant was municipal point sources.

1994

Of the total 34 388 square miles, 26 847 square miles were assessed. Fifteen States reported that extra nutrients pollute 4 548 square miles of estuarine waters, (which equals 47% of the impaired estuarine waters).

1996

Of the total 39 839 square miles of estuaries, the 28 819 square miles were surveyed. The States identified more square miles of estuarine waters polluted by nutrients than any other pollutant or process (Figure 13). Eleven States reported that extra nutrients pollute 6,254 square miles of estuarine waters (57% of the impaired estuarine waters). The top three ranked sources of pollution for estuaries were Industrial Discharges (21% of surveyed waters) Municipal Point Sources (18%) and Urban Runoff/Storm Sewers (17%).

Only 6% of shoreline waters were assessed.

1998

Assessed Waters Total estuaries = 90 465 square miles a Total assessed = 28 687 square miles

10% of assessed miles-23% of impaired miles due to nutrients-municipal point sources are ranked first in the list of stressors.

The states also report that excess nutrients impact 2 880 square miles (10% of the assessed estuarine waters and 23% of the impaired estuarine waters).

GREAT LAKES

1992

Information for 1992 is limited but the main polluters were reported as atmospheric depositions and contaminated sediments.

1994

Of the total of 5 559 miles, 5 224 miles or 96% were surveyed. The largest source of pollution being Priority Toxic Organic accounting for 98%

Nutrients are ranked fourth behind toxic organic chemicals and pesticides.

1996

Nutrients are ranked fourth as a source of pollution for the Great Lakes, the main source of pollution being atmospheric depositions.

1998

Of the shoreline: 5 521 4 950 miles 90% were assessed, 96% of which was impaired. 5% of the assessed shoreline was reported as impaired by nutrients and 5% of total shoreline assessed was also impaired by nutrients.

SUMMARY TABLES

1992

Water body type	% assessed	% of impaired due to nutrient	% of assessed that is nutrient impaired	% of impaired due to agricultural run-off	% of impaired due to municipal waste	% of eutrophic lakes
Rivers	18	37	18*	72*	15	
Lakes	46	40	NR	56	21	32
Estuaries		55				
Great Lakes	99	5	NR	NR	NR	NR

* = Note: Percentages do not add up to 100% because more than one pollutant or source may impair a river segment.

NR = Not Reported

1994

Water body type	% assessed	% of impaired due to nutrient	% of assessed that is nutrient impaired	% of impaired due to agricultural run-off	% of impaired due to municipal waste	% of eutrophic lakes
Rivers	17	24	8	60	17	NA
Lakes	42	43	16	50	19	36
Estuaries	78	47	17	34	39	
Great Lakes	94	6		4		

1996

Water body type	% assessed	% nutrient impaired	% impaired due to agricultural run-off	% impaired due to municipal waste	% of eutrophic lakes
Streams	19	14	25	5	na
Lakes	20	20	19	7	36
estuaries	72	22	10	17	na
Great Lakes 5 186 – surveyed	94%	6			

1996- The 5 Leading Causes of Water Quality Impairment

Rank	Rivers	Lakes	Estuaries
1	Siltation	Nutrients	Nutrients
2	Nutrients	Metals	Bacteria
3	Bacteria	Siltation	Priority Toxic
4	Oxygen-Depleting Substances	Oxygen-Depleting	Oxygen-Depleting Substances
5	Pesticide	Noxious Aquatic Plants	Solids Oil and Grease

1998

Water body type	% assessed	% of impaired due to nutrient	% of assessed that is nutrient impaired	% of impaired due to agricultural run-off	% of impaired due to municipal waste	% of eutrophic lakes
Streams	23	29	10	60	35	NA
Lakes	42	20	44	30	11	38
estuaries	32	23	10	15	28	NA
Great lakes	90	5	5	3		

APPENDIX E COST AND ENERGY MODEL OF WASTEWATER AND SLUDGE TREATMENT

A spreadsheet model was constructed to calculate annual costs and energy used by wastewater and sludge treatment processes. This was derived from existing models used by WRc for several years. Three wastewater treatment plants were modelled (table E.1).

Table E.1 Wastewater and sludge treatment processes modelled

Model	PE	Wastewater treatment	Sludge treatment & disposal
1A	20000	Secondary activated sludge with denitrification, chemical P removal	Digestion ¹ , dewatering ¹ and application of sludge to agricultural land
1B	20000	Secondary activated sludge with denitrification, chemical P removal	Incineration ¹
1C	20000	Secondary activated sludge with denitrification, chemical P removal	Digestion ¹ , dewatering ¹ and landfill
2A	200000	Secondary activated sludge with denitrification, biological P removal with chemical backup	Digestion, dewatering and application of sludge to agricultural land
2B	200000	Secondary activated sludge with denitrification, biological P removal with chemical backup	Incineration ¹
2C	200000	Secondary activated sludge with denitrification, biological P removal with chemical backup	Digestion, dewatering and landfill

Note 1. After transport to a larger sludge treatment centre

The spreadsheet calculates an annual cost as the sum of the operating cost and an annualised investment cost, assuming a 20 year life for mechanical assets and a 40 year life for civil assets.

All significant costs and energy inputs or outputs are taken into account: staff, chemicals, fuel, power, transport, sludge digestion, incineration, the value of re-used phosphorus and nitrogen in sludge applied to land. The main assumptions are shown in table E.2.

Table E.2 Process model assumptions

<i>Phosphorus removal</i>	
Primary settlement	15%
Secondary biological treatment	20%
Extra in biological phosphorus removal	50%
Chemical phosphorus removal	Fe:P ratio of 1.5 to 1 is sufficient to meet standard
<i>Value of re-used phosphorus</i>	
Price (euro/tonne P)	132
Energy (GJ/tonne P)	28
<i>Phosphorus utilisation</i>	
In sludge put on agricultural land	30%
Recovered as side stream	70%
Incineration	0%

The ways in which the model has been used are described in the main report in sections 7. Here the main results are tabulated. In these tables the sensitivity of the outputs to different assumptions is shown:

- Range of P concentrations in crude sewage (8 to 15 mg/l, tables E.3 to E.5), corresponding to different proportions of detergent built from STPP (0% to 100%).
- Phosphorus availability: 50% in sludge applied to land (table E.6), and 50% of the sidestream phosphorus (table E.7).

In some countries, less than 100% of the area is (or is expected to be) classed as sensitive. Phosphorus discharges to just sensitive areas have therefore been estimated, making some broad assumptions (table E.8).

Table E.3 Process model results, 12 mg/l P in crude sewage

Model	Description	PE	Annual cost	NPE	P in crude sewage	P in effluent	P in sludge	P in sidestream	Available P re-used	P lost
			Euro/y/pe	GJ/y/pe	t/year	t/year	t/year	t/year	t/year	t/year
1A	Chemical P removal Sludge to land	20000	19.36	0.22	27.6	3.4	24.1		4.8	22.8
1B	Chemical P removal Sludge incineration	20000	22.27	0.43	27.6	3.4	24.1		0.0	27.6
1C	Chemical P removal Sludge to landfill	20000								
2A	Biological P removal with chemical backup Sludge to land	200000	6.25	0.11	275.9	11.5	264.4		132.2	143.7
2B	Biological P removal with chemical backup Sludge incineration	200000	11.00	0.30	275.9	11.5	264.4		0.0	275.9
2C	Biological P removal with chemical backup Sludge to landfill	200000								

Table E.4 Process model results, 8 mg/l P in crude sewage

Model	Description	PE	Annual cost	NPE	P in crude sewage	P in effluent	P in sludge	P in sidestream	Available P re-used	P lost
			Euro/y/pe	GJ/y/pe	t/year	t/year	t/year	t/year	t/year	t/year
1A	Chemical P removal Sludge to land	20000	18.53	0.19	18.4	3.4	14.9		3.0	15.4
1B	Chemical P removal Sludge incineration	20000	21.13	0.37	18.4	3.4	14.9		0.0	18.4
1C	Chemical P removal Sludge to landfill	20000								
2A	Biological P removal with chemical backup Sludge to land	200000	6.11	0.11	184.0	11.5	172.5		86.2	97.7
2B	Biological P removal with chemical backup Sludge incineration	200000	10.71	0.28	184.0	11.5	172.5		0.0	184.0
2C	Biological P removal with chemical backup Sludge to landfill	200000								

Table E.5 Process model results, 15 mg/l P in crude sewage

Model	Description	PE	Annual cost	NPE	P in crude sewage	P in effluent	P in sludge	P in sidestream	Available P re-used	P lost
			Euro/y/pe	GJ/y/pe	t/year	t/year	t/year	t/year	t/year	t/year
1A	Chemical P removal Sludge to land	20000	19.98	0.23	34.5	3.4	31.0		6.2	28.3
1B	Chemical P removal Sludge incineration	20000	23.12	0.47	34.5	3.4	31.0		0.0	34.5
1C	Chemical P removal Sludge to landfill	20000								
2A	Biological P removal with chemical backup Sludge to land	200000	6.36	0.11	344.9	11.5	333.4		166.7	178.2
2B	Biological P removal with chemical backup Sludge incineration	200000	10.63	0.27	344.9	11.5	333.4		0.0	344.9
2C	Biological P removal with chemical backup Sludge to landfill	200000								

Table E.6 Process model results, 12 mg/l P in crude sewage, P availability in sludge 50%

Model	Description	PE	Annual cost	NPE	P in crude sewage	P in effluent	P in sludge	P in sidestream	Available P re-used	P lost
			Euro/y/pe	GJ/y/pe	t/year	t/year	t/year	t/year	t/year	t/year
1A	Chemical P removal Sludge to land	20000	18.24	0.16	27.6	3.4	24.1		12.1	15.5
1B	Chemical P removal Sludge incineration	20000	20.60	0.29	27.6	3.4	24.1		0.0	27.6
1C	Chemical P removal Sludge to landfill	20000								
2A	Biological P removal with chemical backup Sludge to land	200000	6.06	0.12	275.9	11.5	264.4		132.2	143.7
2B	Biological P removal with chemical backup Sludge incineration	200000	10.55	0.26	275.9	11.5	264.4		0.0	275.9
2C	Biological P removal with chemical backup Sludge to landfill	200000								

Table E.7 Process model results, 12 mg/l P in crude sewage, sidestream P availability 50%

Model	Description	PE	Annual cost	NPE	P in crude sewage	P in effluent	P in sludge	P in sidestream	Available P re-used	P lost
			Euro/y/pe	GJ/y/pe	t/year	t/year	t/year	t/year	t/year	t/year
1A	Chemical P removal Sludge to land	20000	18.24	0.17	27.6	3.4	24.1		7.2	20.4
1B	Chemical P removal Sludge incineration	20000	20.60	0.29	27.6	3.4	24.1		0.0	27.6
1C	Chemical P removal Sludge to landfill	20000								
2A	Biological P removal with chemical backup Sludge to land	200000	6.06	0.13	275.9	11.5	264.4		79.3	196.6
2B	Biological P removal with chemical backup Sludge incineration	200000	10.55	0.26	275.9	11.5	264.4		0.0	275.9
2C	Biological P removal with chemical backup Sludge to landfill	200000								

APPENDIX F SLUDGE PRODUCTION ESTIMATES

Sludge production is highly dependent on the crude sewage characteristics and the sewage treatment processes.

	Influential factors
Crude sewage	COD/BOD concentration Suspended solids Soluble P concentration – if chemical P removal
Processes	Rate of biological treatment – less sludge from denitrifying processes Chemical P removal generates sludge – <i>the dose of chemical is critical</i>

WRc estimates of sludge production g/pe/day. See below for details.

Treatment process	a) 100% STPP	b) 100% Zeolite A	c) 50% STPP
AS, no P removal	58	65	61.5
AS, chemical P removal	82	78	80
AS, biological P removal, chemical back up	66	61	63

Cases considered

Processes

1. Activated sludge without nutrient removal.
2. Activated sludge with denitrification and chemical P removal.
3. Activated sludge with denitrification and biological P removal. Chemical P removal used as back up.

Detergent builders

- a) Detergents 100% built from STPP
- b) Detergents 100% built from Zeolite A
- c) Intermediate cases by interpolation

All calculations are per population equivalent

Detailed calculations

Phosphorus based detergents

Human and food waste	1.8 g/pe/day (ref 1)
In STPP if 100% laundry & dishwasher detergent is STPP based	8 kg detergent/person/year STPP is 24% of detergent P is 25% of STPP $8000 * 0.24 * 0.25 / 365$ g/person/year = 1.3 g/pe/day

Zeolite based detergents

Human and food waste	1.8 g/pe/day
If 100% laundry detergent is Zeolite A based Assume dishwasher detergent is still phosphate based	6.4 kg laundry detergent/person/year 1.6 kg dishwasher detergent, assume still STPP based <i>Zeolite A</i> (25% of detergent): $6400 * 0.25 / 365$ g/person/year = 4.4 g/pe/day <i>PCAs</i> and carbonate designed to precipitate Ca & Mg will add to sludge quantity, relative to STPP based detergents. Difficult to estimate the quantity precisely, say add 50-60%, making total of 7 g/pe/day sludge generated. See table 2.2 of the draft report for detergent contents. <i>P</i> : $1600 * 0.24 * 0.25 / 365$ g/person/year = 0.26

P per day in crude sewage is (a) 3.3 g/pe/day or (b) 2.0 g/pe/day (rounded down from 2.06)

Primary settlement

- Assume sludge quantity 27 g/pe/day. Based on 60% removal of solids, 150 mg/l SS in crude sewage, 300 l/pe/day (these are median values from a benchmarking survey carried out by WRc in 2001).
- P removal is 15 to 30%. Say 25%
- P remaining after primary settlement is 2.48 g/pe/day (a) or 1.5 g/pe/day (b)

Activated sludge

Assume 60 gBOD/pe/day in crude sewage (consistent with ref 1 and WRc benchmarking study)

Assume 40% removed in primary sludge = 24 g/pe/day

Assume 3 g/pe/day discharged in treated effluent

Therefore 33 g/pe/day is oxidised in activated sludge

Assume 0.8 to 1.1 kg sludge / kgBOD removed (higher than 0.62 = 0.31 per kg COD assumed in ref 1)

Therefore, without denitrification, biological sludge production is 26.4 to 36.3 g /pe/day. Typically 31 g/pe/day.

Activated sludge with denitrification – longer sludge residence time – smaller quantity of sludge. Assume 26.4 g/pe/day

Assume AS by itself removes a further 15% P (of crude sewage content), so that total removal by standard primary settlement and activated sludge is 40%.

P in treated effluent

Assume 0.73 mg/l, or 0.22 g/pe/day at 300 l/pe/day

Chemical P removal

Assume ferric chloride (Etienne ref 1).

To estimate the quantity of ferric chloride, use a molar ratio P:Fe. The P is the extra above what would be removed anyway through activated sludge (ref 2 and ref 1.). Ref 1 indicates that a ratio of 1.5 may be adequate. Refs 3 and 4 suggest that 2.5 might be needed, and ref 2 shows that the Fe dose required to achieve a soluble P residual less than 1 mg/l can be quite high. Look at range 1.5/2.0/2.5.

Sludge production is 6.6/8.3/10.0 g / g P removed chemically (ref 1)

P to be removed chemically is (a) $(0.6 * 3.3 - 0.22) = 1.76$ g/pe/day or (b) $(0.6 * 2.0 - 0.22) = 0.98$ g/pe/day, without biological P removal. Sludge production is (a) 11.6/14.6/17.6 g/pe/day or (b) 6.5/8.1/9.8 g/pe/day

Biological P removal (BPR)

Assume it is capable of removing enough so that in case b) no chemical treatment is needed to achieve <0.8 mg/l. At 300 l/pe/day (includes some rainwater), this is 0.22 g/pe/day. Therefore BPR removes 1.0 g/pe/day.

With biological P removal, the extra is (a) 0.76 g/pe/day or (b) 0.0 g/pe/day, and sludge production is (a) 5.0/6.3/7.6 g/pe/day or (b) 0.0 g/pe/day.

Notes:

Chemicals. Ref 1 shows that approx. 70% of French processes use ferric chloride. Churchley (ref 2): various salts including ferrous and ferric chloride and sulphate. Aluminium salts are more expensive and are used in a minority of cases.

Emschergenossenschaft (personal communication) normally use ferrous or ferric chloride.

Dosing is commonly done after primary settlement and before biological treatment, where less chemical is needed than dosing before primary settlement.

Calculated sludge production (assuming Fe:P ratio of 2:1, method is the same for other molar ratios)

1) Activated sludge without nutrient removal

a) 100% STPP

– Primary sludge	27 g/pe/day
– Secondary sludge	31 g/pe/day
– Extra for P removal	0 g/pe/day
– Extra due to Zeolite A etc	0 g/pe/day
– Total	58 g/pe/day

b) 100% Zeolite A

– Primary sludge	27 g/pe/day
– Secondary sludge	31 g/pe/day
– Extra for P removal	0 g/pe/day
– Extra due to Zeolite A etc	7 g/pe/day
– Total	65 g/pe/day

2) Activated sludge with chemical P removal

a) 100% STPP

- Primary sludge 27 g/pe/day
- Secondary sludge 31 g/pe/day
- Extra for P removal 15 g/pe/day
- Extra due to Zeolite A etc 0 g/pe/day
- Total 73 g/pe/day (+26% compared to 1a)

b) 100% Zeolite A

- Primary sludge 27 g/pe/day
- Secondary sludge 31 g/pe/day
- Extra for P removal 8 g/pe/day
- Extra due to Zeolite A etc 7 g/pe/day
- Total 73 g/pe/day

3) Activated sludge, biological P removal with chemical back up

a) 100% STPP

- Primary sludge 27 g/pe/day
- Secondary sludge 26 g/pe/day
- Extra for chem P removal 8 g/pe/day
- Extra for bio P removal 1 g/pe/day
- Extra due to Zeolite A etc 0 g/pe/day
- Total 62 g/pe/day

b) 100% Zeolite A

- Primary sludge 27 g/pe/day
- Secondary sludge 26 g/pe/day
- Extra for chem P removal 0 g/pe/day
- Extra for bio P removal 1 g/pe/day

- Extra due to Zeolite A etc 7 g/pe/day
- Total 61 g/pe/day

Summary table, calculations modified by experience

Estimated sludge production (g/pe/day)

Treatment process	a) 100% STPP	b) 100% Zeolite A	c) 50% STPP
AS, no P removal	[58]	[65]	[61.5]
AS, chemical P removal	73 (76) [82]	73 (75) [78]	73 (75.5) [80]
AS, biological P removal, chemical back up	62 (63) [66]	61 (61) [61]	61.5 (62) [63]

Assuming Fe:P molar ratio of 2 (2.5). The figures in [] are estimated from UK experience (50% STPP) that adding chemical P removal adds ~30% to the quantity of sludge. This corresponds to extra sludge 1.6* the estimates derived from a molar ratio of 2.

The extra *quantities* agree with those estimated by Etienne et al. The proportions do not, because Etienne et al assume a very low quantity of sludge produced (<40g/pe/day).

Churchley (personal communication) states that Severn Trent Water assume 30% extra sludge as a working assumption. Bill Storey (Water Service Northern Ireland, personal communication) indicates 20-40% extra sludge for the Lough Neagh treatment works. This is for the UK, where approx. 50% of detergents are STPP based. This would imply 80 g/pe/day. The difference between this and the theoretical estimates may be due to higher dosing rates than assumed (molar ratio >~3?) – adopted to be sure of meeting limits on P in the effluent. The tendency to dose generously would apply to cases 2 a, b and c and 3 a and possibly c.

The figures in square brackets [] have been used in the main report. They indicate that, given a mix of sewage treatment without P removal and with P removal, the choice of detergent builder has negligible influence on the quantity of sludge generated.

References

1. Etienne et al, Excess Sludge Production and Costs due to Phosphorus Removal, paper given at NoordWijkerhoot Conference on P recovery, 2001.
2. Experiences with chemical phosphorus removal, John Churchley, in Aqua-Enviro Conference on the Removal of Phosphorus and Recovery from Sludge, Leeds UK, October 2001.
3. Metcalf and Eddy, Wastewater Engineering, McGraw Hill.
4. Siegrist and Boller, Effects of the Phosphate Ban on Sewage Treatment, EAWAG news 42, July 1997

APPENDIX G ZEOLITE A

The potential of zeolites to remove nutrients such as N and P and heavy metals has been recognised for a number of years. Zeolite A is a synthetic negatively charged crystalline aluminosilicate consisting of a three dimensional array of aluminosilicate tetrahedra, which form a honeycomb structure (Douglas and Coad 1997, Dwyer *et al* 1990).

Zeolite A softens wash water by sequestering cations such as Na⁺, K⁺, Ca⁺ and Mg⁺ by ion exchange. As cavities within zeolites constitute a high percentage of their volume, the internal surface area is much larger than the external surface area, and therefore has a large capacity for ion exchange compared to other aluminosilicate minerals. Extensive studies have shown the ability of Zeolite A to bind a variety of metals from solution (Allen *et al*, 1983). The affinity of metal adsorption and release is influenced by factors such as water hardness, pH and temperature (Dwyer *et al*. 1990). Zeolite A has been demonstrated to hydrolyse extensively in natural waters, with a half-life of approximately 1-2 months (Douglas and Coad 1997). The rate of hydrolysis increases with decreasing pH. Zeolite A is thought to be completely soluble at a pH of <3.

Due to its molecular sieve properties, concerns have been expressed that the degradation and hydrolysis of Zeolite A may allow adsorbed metals and other objectionable organics to be remobilised into receiving waters. However, Allen *et al* (1983) showed that metal exchange for cadmium, copper, nickel and zinc was less than 10% (but higher for lead). This exchange rate increased at higher Zeolite A concentrations. Subsequently, Morse *et al* (1994) concluded that while the concern regarding the interaction with Zeolite A and metal exchange is minimal, the concern is not zero, and further investigations may be warranted in areas with inadequate waste water treatment or elevated metal concentrations.

Dissolved silica and aluminium may be released during the degradation of Zeolite A. The release of dissolved silica has the potential to alter the community structure phytoplankton populations and promote the growth of diatoms, who utilise silica in frustule development. Concerns related to the release of dissolved aluminium are related aluminium toxicity to fish under low pH conditions, and the link between aluminium and Alzheimers disease (Baker and Schofield 1981, Morse *et al* 1994). However, this is expected to be low due to the low relative contribution of aluminium from Zeolite A, compared to other sources (Morse *et al* 1994).

Studies on Zeolite A have shown that it is non toxic at projected environmental levels to freshwater and marine species and does not contribute to the eutrophication potential of surface waters (Maki and Macek 1978). Overall, Zeolite A is thought to be only slightly toxic to mammals and has no acute or chronic toxicity to humans via dermal, ocular and respiratory routes of exposure (Dwyer *et al* 1990, Douglas and Coad 1997).

A review by Dwyer *et al* 1990 showed up to 90% of Zeolite A is removed during the activated sludge process, and this removal was not directly related to influent concentrations. It was found that removal rates appeared to be unaffected by influent concentrations under all treatment conditions, which was presumed to result from Zeolite As insolubility and hence removal by sedimentation.

References

Allen, H.E., Cho, S.H. and Neubecker T.A. (1983). Ion Exchange and Hydrolysis of Type A Zeolite in Natural Waters. *Water Res.*, (17), 1871-1879.

Baker J., and Schofield C. (1982). Aluminium Toxicity to Fish in Acidic waters. *Water, Air and Soil Pollution*, (18), 289-309.

Douglas, G.B. and Coad, D.N. (1997). Review of estuarine sediment remediation techniques. Confidential Report prepared for Water and Rivers Commission. CSIRO Division of Water Resources Report (96-11). Revised October 1997. 96pp.

Dwyer M., Yeoman S., Lester J., and Perry R (1990). A Review of Proposed Non-Phosphate Detergent Builders, Utilisation and Environmental Assessment. *Environmental Technology.*, (11), 263-294

Maki A., and Macek K., (1978). Aquatic Environmental Safety Assessment for a Nonphosphate Detergent Builder. *American Chemical Society*, (12), 573-580.

Morse G., Lester J., and Perry R. (1994). The Environmental and Economic Impact of Key Detergent Builder Systems in the European Union. Environmental Engineering Laboratory, Imperial College of London, Technology and Medicine, London SW7 2BU.