50 years of air quality control in Northwestern Germany – how the blue skies over the Ruhr district were achieved

Part II

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Abstract Part II of our short history of air pollution control in Northwestern Germany covers the period from 1975 to 2000. The creation of a comprehensive federal legislation, complemented by initiatives from the states, is described. European legislation played a minor role at that time, with the exception of traffic exhaust control. The effective implementation of the new legislation by the competent authorities of the federal states in Germany and new elements such as regional air quality plans, giving rise to a fast decrease of industrial emissions, are outlined. Emissions from other sectors such as traffic are quantified. The resulting trends of major pollutants in ambient air are highlighted, demonstrating a net improvement of air quality.

50 Jahre Luftreinhaltung in Nordwest-Deutschland – wie der blaue Himmel über der Ruhr zurückgewonnen wurde – Teil II


1 Introduction

Around 1975, the legal foundations of a modern clean air policy had been established in Western Germany, particularly by the Federal Immission Control Act [1] and the first update of the Technical Instructions on Air Quality Control [2] from 1974. However, the mission of reducing pollution levels was far from completed. Whereas the first decade of intensive air quality control from 1960 to 1970 focussed on the reduction of dust emissions (see Part I [3]), sulfur dioxide emissions stayed more or less constant and nitrous oxide emissions were on the rise. Important noxious compounds such as heavy metals and other constituents of suspended particulates (TSP) or benzene were not regulated at all, partly because of the limitations in the analytical techniques to monitor them in flue gases or in ambient air at that time. Secondary air pollutants, for example ozone, were widely unknown, let alone highly toxic compounds such as dioxins or furans. Part II of our short history of air pollution control covering the period from 1975 to 2000 will describe which strategies were developed to tackle these problems. Basically the list of substances of particular relevance for the environment was extended and stringent emission limits based on the precautionary principle were established. As in the first part [3], we will consider the development of environmental legislation (section 2) in parallel with the resulting abatement measures (section 5) and their impact on emissions (section 4) and ambient air quality (section 5). The limited scope of the article at hand forbids a detailed description of all legislation or data. Instead, we will focus on the major trends. For more details the reader will be referred to the literature.

2 Extending and sharpening the legal instruments

2.1 Federal and state level

One of the characteristics of the period from 1975 to 2000 is a formidable extension of the environmental legislation. The basis of air pollution control in Germany, the Federal Immission Control Act [1], has repeatedly been amended, however without changing its essence. An overview is given in [4]. Particularly the updates in 1985 [5] and 1990 [6] were important. In 1985, the economic appropriateness as prerequisite to improve flue gas cleaning was replaced by the general principle of proportionality [4], opening the way to a systematic update of industrial facilities in the Technical Instructions on Air Quality Control from 1986 [7] (see below). In 1990 [6], regulations for plant safety (beyond the scope of this article) and for traffic restrictions irrespective of smog episodes (article 40, paragraph 2) were added. Based on the Federal Immission Control Act, 28 ordinances had been issued by 1999 [8], covering certain types of installations or regulating specific aspects such as the authorization procedure. An overview of ordinances with relevance for this article is presented in Table 1. The 15th ordinance [9] was such an important milestone that it deserves a particular mention.

It finally paved the way for the general flue gas desulfurization of large combustion plants by setting an emission limit of 400 mg/m³ for SO₂ for plants burning solid fuels with a capacity above 500 MW [9]. This was the endpoint of a fierce struggle between the competent authorities and industry. Already in 1974, flue gas desulfurization was technically available and more than 50 units were operating or under construction in the United States and Japan [18]. However, industry claimed the costs were disproportionate, particularly for existing plants, so that flue gas desulfurization had to be implemented against strong resistance [19]. It is questionable whether this final success for the precautionary principle would have been possible without the upcoming debate on the observed new forest decline [20], which was at
The 13th ordinance did not prescribe ambitious emission limits for nitric oxides (800 mg/m³ for large combustion plants burning solid fuels), which would have required a denitrification unit. Here the German states came into play. In April 1984, a conference of the ministers for the environment of the German states declared an emission limit of 200 mg/m³ for nitric oxides as best available technique and as general requirement for the authorization, opening the way also for denitrification units [21].

The updates of the “Technical Instructions on Air Quality Control” from 1974 [2], 1985 [22] and 1986 [7] were important milestones on the road to clean air. The update from 1974 [2] extended the list of compounds with emission limit values considerably, in particular by including noxious constituents of dust such as arsenic, lead or cadmium compounds or by emission standards for three classes of organic compounds (for example benzene was classified into class 1 with an emission limit of 20 mg/m³). Setting emission standards for carcinogenic compounds as well was a specification of the general overriding principle to keep the emissions of carcinogens as low as possible. The emissions of certain compounds such as SO\(_2\), NO\(_x\), or Cl\(_2\) had to be monitored continuously, if certain emission fluxes were exceeded. In addition to the general regulations, emission limit values specific for certain industrial installations were introduced. Ambient air quality standards were set for Cl\(_2\), HCl, HF, CO, SO\(_2\), H\(_2\)S, NO\(_2\) and NO (see also table 5 in Part I [3]).

The update from 1985 related to the lowering of ambient air quality standards (see table 5 of Part I [3]) and to the introduction of new standards such as lead and cadmium compounds in total suspended particulates (2 µg/m³ and 0.04 µg/m³, respectively).

Of great importance was the update in 1986 [7] by setting new stringent emission standards based on the precautionary principle and in particular by introducing the requirement of refurbishing existing facilities within five years as a general rule (for a detailed description of the updates see [23]). The specification of best available techniques by setting emission standards could draw on technical guidelines issued by the Federation of German Engineers [24].

Table 1. Important ordinances based on the Federal Immission Control Act.

<table>
<thead>
<tr>
<th>Ordinance, year</th>
<th>Subject</th>
<th>Commentary</th>
</tr>
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<tbody>
<tr>
<td>1st [13], 1988</td>
<td>Small combustion plants</td>
<td>Replaced preceding ordinances of German states, update in 1997 [16]</td>
</tr>
<tr>
<td>13th [9], 1983</td>
<td>Large combustion plants</td>
<td>Stringent emission limits for SO(_2)</td>
</tr>
<tr>
<td>17th [10], 1990</td>
<td>Incinerators</td>
<td>Stringent emission limit for dioxins/furans, partly motivated by public resistance against municipal incinerators</td>
</tr>
<tr>
<td>20th [11], 1998</td>
<td>Limiting VOC emissions from storage of gasoline</td>
<td>Important for reducing ozone peak concentrations</td>
</tr>
<tr>
<td>21st [12], 1992</td>
<td>Limiting VOC emissions from fuelling of vehicles</td>
<td>Important for reducing ozone peak concentrations</td>
</tr>
<tr>
<td>22nd [14], 1993</td>
<td>Transposition of European air quality limit values into German law</td>
<td>Enforced by European court rulings [17], that transposition in a technical instruction was not sufficient</td>
</tr>
<tr>
<td>23rd [15], 1996</td>
<td>Traffic restrictions irrespective of smog episodes</td>
<td>Thresholds for NO(_2), soot (black carbon), benzene</td>
</tr>
</tbody>
</table>

Taking the increasing importance of traffic on ambient air quality into account, the extension of the environmental regulations did not only refer to industrial installations, but also covered the quality of fuels, for example by restricting the content of lead or benzene in gasoline.

An early example is the leaded fuel act from 1971 [25], limiting the lead content in two stages to 0.40 g/l and 0.15 g/l, respectively. More than a decade later, product standards for fuels including lead, benzene and unleaded gasoline were set in ordinances, as in 1988 [26] and in 1993 [27] as 10th ordinance to the Federal Immission Control Act [1; 5; 6]. These ordinances were also used to transpose European into national law (see section 2.2). Of importance for the control of highly toxic chlorinated compounds (see section 5) was legislation in adjacent environmental fields such as chemical safety. The use of certain toxic compounds, for example polychlorinated biphenyls (PCB) or vinyl chloride (VC), was first restricted (1978, [28]) and later prohibited (1989, [29]).

The most important input by the German states, the adoption of a strict emission standard for NO\(_x\), [21], has already been mentioned. In North Rhine-Westphalia, the State Act on Air Pollution Control was amended in February 1975 [50], inter alia enabling the authorities to delineate special zones with high pollution loads, where air quality plans had to be established and measures from these plans could be made compulsory [51]. This was the implementation of a federal legal instrument introduced in 1974 by the Federal Immission Control Act [1], enabling regional air quality plans.

Other important achievements on the state level were air quality criteria for carcinogenic air pollutants such as benzene (2.5 µg/m³), benzo[a]pyrene (1.3 ng/m³) or the cadmium content in TSP (1.7 ng/m³) in 1991 [32] and information (180 µg/m³) and alert thresholds (560 µg/m³) for ozone in 1990 [53]. These criteria were not adopted by national law, but were used for special cases in the licensing procedure, as target values for air quality plans (criteria for carcinogenic air pollutants) and for the information of the public during episodes of high ozone.

Most of these initiatives were prepared by the “state committee on immission control (LAI)”, a working group from the federal and state ministries for the environment, safeguard-
ing a trustful cooperation of the federal and state administrations. This committee also played a decisive role in the coordination of an efficient enforcement of environmental legislation by the German states (see section 5).

2.2 European level

The European Clean Air Policy is based from the beginning on three columns: source related regulations, regulation of important products, and air quality criteria [34].

Up to the nineties, regulations on the European level were of minor importance for the German legislation. On the contrary, German regulations often were prototypes for later European directives [34; 35]. Examples are the directive on large combustion plants from 1988 [36], the petrol directives from 1984 [34; 39], or the ozone directive from 1992 [43], TSP, lead and NO\textsubscript{2} were discussed during a symposium in 1981 [62].

Consequently, the decisive breakthroughs to lower the emissions were rather achieved by special programs implementing new legislation such as the ordinance on large combustion plants [9] (see section 3.2). The lack of legal instruments was also the reason why air pollution originating from traffic could not be tackled.

Unwithstanding these shortcomings in relation to abatement measures, the importance of the first regional air quality plans lies in their systematic approach, taking into consideration the whole process from the emissions of air pollutants to their effects on human health or ecosystems. These plans can thus be regarded as forerunners to the current air quality planning (see Part III).

As both instruments, air quality plans and special programs, were run in parallel, it is difficult to separate the emission reductions due to air quality plans as such from those caused by the special programs. The combined effects of the first two generations of air quality plans and the special programs on emissions and ambient air quality were presented by a record of success in 1989 [61].

The strategy for air quality planning was changed in 1992 by extending air quality plans to areas with a potential of a higher pollution burden (areas subject to investigation) in the whole state of North Rhine-Westphalia (3rd generation), following a corresponding amendment of the Federal Immission Control Act in 1990 (article 44) [6].

3 Abatement Measures in North Rhine-Westphalia

3.1 Air quality plans

Whereas the first systematic programs to abate air pollution focussed on specific industrial branches (see section 2.5 of part I [5]), the strategy was changed in 1975 by introducing comprehensive regional air quality plans for five areas with a particularly high pollution load. These areas comprised large parts of the Rhine-Ruhr district (5,150 km\textsuperscript{2}) with nearly 6 million inhabitants. An overview of the regional air quality plans and their updates is given in Table 2.

These air quality plans contained already the same elements as modern plans such as an emission inventory, a description of the pollution load, source apportionment, modelling (though in a limited form), a forecast and a chapter on abatement measures. Following the fast development of monitoring techniques, many air pollutants were covered (see section 5). In addition, studies on the impact of air pollutants on human health, ecosystems, materials or the soil were presented.

The chapter on abatement measures comprised about 300 to 400 single actions relating to industrial sources, such as the desulfurization of cokery gas or the enclosure of stock piles of raw materials. However, the measures were limited by the legal possibilities to enforce flue gas cleaning (see section 2). Consequently, the decisive breakthroughs to lower the emissions were achieved by special programs implementing new legislation such as the ordinance on large combustion plants [9] (see section 3.2). The lack of legal instruments was also the reason why air pollution originating from traffic could not be tackled.

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3.2 Special abatement programs

As already mentioned in section 2, there was strong resistance against flue gas desulfurization from industry, which was the main reason for the more or less constant SO\textsubscript{2} emissions from 1960 to 1985. In order to reduce the still elevated ambient SO\textsubscript{2} levels, tall stacks up to 500 m were constructed for some power plants and industrial installations such as sintering plants in North Rhine-Westphalia [62]. The better dispersion conditions improved the air quality in the Ruhr conurbation (see Figure 5 in Part I), but enhanced the share to long range transport and gave rise to a larger input of SO\textsubscript{2} and NO\textsubscript{2} into ecosystems. The pros and cons of tall stacks were discussed during a symposium in 1981 [62].
The debate on the observed new types of damages to forests [20] showed that better dispersion by high chimneys was not the solution for a sustainable development, but that the emissions of SO\textsubscript{2} and NO\textsubscript{x} and their input into ecosystems had to be reduced. After the 15\textsuperscript{th} ordinance on large combustion plants [9] and the declaration of the state ministers of the environment [21] on NO\textsubscript{x} limit values had paved the way (see section 2), the competent authorities in North Rhine-Westphalia established an ambitious program for emission reduction at large combustion plants in November 1984 [65]. In the framework of this program, about 100 power station units with an overall capacity of 61,000 MW were retrofitted with flue gas desulfurization in the short period from 1984 to 1989. The achieved annual reduction of SO\textsubscript{2} emissions was 865,000 t (see Figure 1) [65]. Likewise the NO\textsubscript{x} emissions were effectively reduced. Whereas emissions lower than 200 mg/m\textsuperscript{3} (solid fuels) and lower than 100 mg/m\textsuperscript{3} (gas) could already be achieved by primary measures for combustion plants with lignite or gas as fuels, large power plants burning coal (and two units burning oil) had to be retrofitted with catalytic denitrification. By 1990, more than 65 power plant units with an overall capacity of 40,000 MW were equipped with catalytic denitrification. NO\textsubscript{x} emissions could be reduced by 275,000 tons per year [65]. Flue gas desulfurization and denitrification also lowered the annual dust emissions by 55,000 tons [65] (see Figure 1).

This program was a big success of the competent authorities, the factory inspectorates, owing to an efficient and fast implementation of the new legal instruments. The next systematic abatement program was based on the update of the “Technical Instructions on Air Quality Control” from 1986 [7], opening the way for modernizing existing facilities within five years as a general rule. About 8,900 industrial installations had to be checked, from which about 5,900 installations had to be refitted [64]. A task force to implement this program was established. In 1991, already 70\% of the facilities had been modernized [65], and in 1992, this percentage rose to 78\%, including installations with longer deadlines up to 1996 [64].

Likewise the systematic program to reduce the emissions of polychlorinated dioxins and furans (PCDD/PCDF) from waste incinerators, starting in 1990, was an efficient implementation of new federal law, in this case the 17\textsuperscript{th} ordinance on incineration plants [10], setting an ambitious limit value of 0.1 ng/m\textsuperscript{3} for PCDD and PCDF (the sum of international toxicity equivalents [67]). 26 waste incinerators were investigated [65] in order to meet the new emission standard as early as possible, partly using voluntary agreements. By 1993, six installations already met the emission limit, and eleven installations were in the process of refitting, using either active charcoal filters or injection, catalytic destruction of PCDD/PCDF or a combination of these techniques with primary measures such as high combustion temperatures [68].

Whereas around 1990 high dioxin emissions by waste incinerators and by the production of certain organochlorine compounds such as trichlorophenol were well known [69], there was high uncertainty on the annual emissions by other industrial processes [70; 71], giving rise to estimates varying from 58 to 580 g toxic equivalents for Germany [72]. This was the reason for a systematic monitoring program of dioxin emissions from 42 industrial installations, starting in 1992. High dioxin emissions and fluxes originating from 14 plants comprising sintering facilities, secondary copper smelters and other recycling installations for metals were discovered [71; 73]. Annual emissions of 500 g toxic equivalents were caused by these installations [73]. These results gave rise to a systematic program to reduce the dioxin and furan emissions from industrial sources aiming to meet an emission limit of 0.1 ng/m\textsuperscript{3}. It took until 2002, before this standard became compulsory on the federal level (see Part III).

### 3.3 Pollution by traffic

Whereas around 1960, the number of vehicles (8 million) in Germany was still rather small [74], this number had more than doubled by 1970 (16.8 million) [74] and steeply increased further to reach 47.5 million vehicles in 1995 [74]. Consequently, this source category was of increasing importance particularly for VOC and NO\textsubscript{x} emissions and indirectly for secondary air pollutants such as ozone. Whereas the implementation of source related federal legislation such as the leaded fuel act [25], the 10\textsuperscript{th}, the 20\textsuperscript{th} and 21\textsuperscript{th} ordinance to the Federal Immission Control Act (see section 2) or the European regulations for exhaust emissions [58] (see Part III) reduced the emissions considerably and caused a net improvement of air quality (see sections 4 and 5), efforts to reduce the impact of traffic at pollution hot spots or to prevent peak concentrations by short term actions for primary and secondary air pollutants remained without success.
The 23rd ordinance [15] in combination with article 40, paragraph 2 of the Federal Immission Control Act [6] enabled the authorities to restrict traffic, if certain thresholds for peak NO\textsubscript{2} concentrations and for benzene and soot annual means were exceeded (for example 14 µg/m\textsuperscript{3} for soot from 1995 and 8 µg/m\textsuperscript{3} from 1998 onwards). However, these thresholds were so high that only few, if any, traffic restrictions could be based upon this ordinance. At least, it gave rise to increasing monitoring and modelling at traffic hot spots (see section 5) and to public awareness.

Art. 40a) of the Federal Immission Control Act [6] opened in 1990 as an additional effort the possibility for short term actions (traffic ban) to reduce peak ozone concentrations exceeding 240 µg/m\textsuperscript{3} (hourly mean at at least three stations). However, field experiments, model calculations and the experience from a summer smog ordinance in the state of Hesse demonstrated the limited potential of regional, let alone local short term actions on peak ozone concentrations [75], due to its formation during transport and not close to a source, which leads to the spatially widespread character of ozone episodes in Western and Central Europe [76].

4 Development of emissions

As discussed in section 3.2, the years from 1983 to 1990 brought the decisive break through for the abatement of industrial SO\textsubscript{2} and NO\textsubscript{x} emissions, particularly from large power plants. The success of these abatement programs is clearly reflected by the trend of SO\textsubscript{2} and NO\textsubscript{x} emissions in North Rhine-Westphalia presented in Figures 2 and 3.

Emissions from industrial sources were calculated from the declarations of emissions according to the 11th ordinance [77] to the Federal Immission Control Act from 1985 onwards every four years and were interpolated for the years in between. For 1977, the emissions were estimated from data published in the air quality plans (see section 3.1) and extrapolated from the five areas with a particularly high pollution load to North Rhine-Westphalia. Emissions from traffic and from small consumers/domestic heating (since 1980) were based on the consumption of fuels in combination with emission factors, which were updated according to the technical progress. VOC emissions from traffic and from solvents including the use of products were converted from German data [78] to North Rhine-Westphalian emissions. These emission data have not been published so far.

Industrial SO\textsubscript{2} emissions decreased between 1983 and 1990 by 90% from about 1.2 to about 0.2 million tons, respectively (Figure 2). Before 1985, the SO\textsubscript{2} emissions had been more or less constant for about two decades (see Part I [3] and Figure 2). Since the emissions from other source categories (traffic and small consumers) were small, industrial emissions dominated the overall trend. A particular steep decrease of SO\textsubscript{2} emissions occurred after 1987, while in the winters of 1985 and 1987, a potential for regional smog episodes (see section 5) was still present.

Industrial NO\textsubscript{x} emissions showed a similar trend, decreasing by more than 70% from about 0.7 million tons in 1985 to about 0.22 million tons in 1990 (Figure 3). In the two decades before, an increase of NO\textsubscript{x} emissions is evident (see Part I [5] and Figure 2). In contrast to the negligible contribution of traffic to overall SO\textsubscript{2} emissions, NO\textsubscript{x} emissions from traffic became the dominant source category from 1990 onwards. Until 1980, NO\textsubscript{x} emissions from traffic had increased, due to the increasing volume of traffic and remained nearly con-
stant in the following decade. Not before the beginning of the nineties a slight decrease by about 20% until 2000 started due to the gradual introduction of three way catalysts in vehicles. Summing up the contributions of all source categories, the decrease of NO\textsubscript{x} emissions was much smaller than the decrease of SO\textsubscript{2} emissions.

As already mentioned in section 3.2, the installation of SO\textsubscript{x} and NO\textsubscript{x} scrubbers in large combustion plants, the upgrade of other industrial sources and structural changes also yielded a further decrease of dust emissions (see Figure 4), whereas the principal reduction of dust emissions had taken place in the two decades before (see Part I [3] section 2.4).

From 1977 to 1997, the industrial emissions of respirable particulate matter (PM\textsubscript{10}) could be further reduced by about 90% (see Figure 4). Figure 4 also shows that PM\textsubscript{10} emissions from small consumers and domestic heating and from traffic (exhaust emissions only) were far from negligible. Whereas tailpipe PM\textsubscript{10} emissions remained nearly constant from 1990 to 2000 (older data are not available), dust emissions from domestic heating and small consumers decreased strongly by 95%, due to the gradual displacement of solid fuels (mostly coal and lignite) by gas, to a lesser extent by oil.

Industrial VOC emissions (including methane) decreased by about 90% from 1977 to 2000 (see Figure 5). From 1990 onwards, VOC data from traffic and solvents (including the use of products) are available as well. In the beginning, these emissions were of comparable magnitude and dominated industrial VOC emissions by far. In the following years, there was a remarkable decrease of traffic VOC emissions, whereas emissions from solvents including the use of products decreased only slightly and became the dominant source category. Inspite of the data gaps, it can be assumed that there was a net decrease of overall VOC emissions in the time period considered. Likewise the emissions of polychlorinated dioxins and furans (PCDD, PCDF) from industrial sources could be reduced by more than an order of magnitude. Whereas industrial emissions were estimated to more than 500 g international toxicity equivalents (TEQ) [67] at the beginning of the nineties [73], the emissions decreased to 156 g TEQ in 1996 and to 15 g TEQ in 2008 [79].

5 Trend of the air quality

As already mentioned in section 2, the period from 1975 onwards can be characterized, inter alia, by a distinct increase in the number of air pollutants which were monitored either in flue gases or in ambient air. In addition to the “classical” air pollutants such as SO\textsubscript{2}, suspended particulate matter (TSP) or dust fall, monitoring was stepwise extended to cover continuous measurements of NO\textsubscript{2}, NO, CO and O\textsubscript{3} since 1977 and discontinuous measurements of toxic constituents of TSP such as lead, cadmium (since 1974) or polycyclic aromatic hydrocarbons (PAH) since 1985 [80]. The discontinuous measurements of TSP and their constituents with high volume samplers originally comprised 71 sites in the Rhine-Ruhr area (see Part I [3], section 2.5). Later, this network was reduced to about 50 stations, arranged in a regular grid with 8 km distance between the stations, generally using the same sites as the continuous monitoring (see below). Benzene monitoring started in 1980 [81] and measurements of PCDD and PCDF in ambient air in 1986 [82]. The extension of monitoring had been enabled by the widespread development of new measurement techniques, giving rise to numerous standardized methods by the German Federation of Engineers.
The extension of the continuous monitoring network was closely related to the air quality plans in five districts with high pollution loads (see section 3.1). In 1978, ten automated stations started their measurements in the western Ruhr district increasing to 66 in 1987 [82]. The continuous monitoring stations were generally arranged in a regular grid with 8 km distance between the adjacent stations with few exceptions, e.g. some rural stations. This measurement planning has the advantage of yielding air pollution levels which are spatially representative for the urban background of an extended area such as the Ruhr district (see section 2.5 of Part I [3]). Consequently, long time series can be calculated from the data although minor changes in the monitoring network occurred. The disadvantage of this approach was that some downtown areas such as Cologne were not included and that hot spots such as street canyons were also not covered.

Basically, there was no systematic monitoring at traffic hot spots until January 1980, when two stations at Düsseldorf and Essen marked the start. Three additional stations placed in street canyons followed in 1992 (Düsseldorf) and 1994 (Hagen). The trend of the average annual SO\textsubscript{2} burden in the Rhine-Ruhr area has already been presented in Figure 5 of Part I [3]. This trend can be regarded as the visualization of the history of air pollution control. After a sharp decrease of the annual means in 1965 and a fluctuation afterwards in the following decade, there was an additional net decrease starting in 1980 and continuing (with a short interruption in 1985) for the next two decades. The decrease of the SO\textsubscript{2} burden from 1980 onwards was at least partly due to the better dispersion conditions of high chimneys (see section 3.1), whereas the desulphurization of the flue gases starting in 1985 (see section 3.2) brought the pollution levels down decisively. After the German reunification at the end of 1989 and the subsequent upgrade or closure of big power plants in Eastern Germany and the Czech Republic, long range transport from Eastern Europe [85], contributing a share of 50 to 60% of the SO\textsubscript{2} burden in the eastern part and 50 to 50% in the western part of the Ruhr district [87]. Other parts of Germany such as Hesse, Lower Saxony and Hamburg were affected as well [85].

The TSP trend presented in Figure 4 of Part I [3] shows a more continuous decline than the SO\textsubscript{2} trend, due to the persisting decrease of dust emissions. The unfavourable dispersion conditions in the winter of 1985 are visible as a corresponding bump in the trend. Around the year 2000, TSP levels amounted to less than 25% of the TSP concentrations at the start of the systematic monitoring.

Noxious and carcinogenic components of TSP such as lead, cadmium and benzo[a]pyrene concentrations decreased even faster, so that the dust became less toxic over time. Figures 6 to 8 show the corresponding trends [90]. All components could be reduced to less than 10% of their original concentrations.

Starting 1989 (traffic stations) and 1995 (rural stations) data can also be compared at different station types. A comparison shows that the concentrations of lead and benzo[a]pyrene were nearly twice as high at traffic stations as in the urban background around 1990, whereas cadmium compounds were not elevated. The high cadmium levels at rural stations in the years 1990 and 1992 are probably monitoring artefacts. All components could be reduced to less than 10% of their original concentrations.

The decrement of lead content in ambient air, particularly the stronger decrease at traffic exposed stations, can partly be ascribed to the gradual limitation of the lead content in gasoline (see section 5.3) up to unleaded gasoline at the end of nineties. The soot content of diesel exhaust with adherent polycyclic aromatic hydrocarbons such as benzo[a]pyrene as tracer was diminished as well. However, the trend of cadmium shows that also the abatement measures at industrial sources (better precipitation of dust) played a decisive role (see section 5.1 and 5.2). In addition, structural changes such as the concentration of the steel industry and the closure of most cokeries came into play.
Abatement measures at municipal incinerators and industrial sources (see section 3.2) were also very successful in lowering the concentrations of dioxins and furans in ambient air by about one order of magnitude since 1988. The trends of these compounds have recently been published [79], and the reader is kindly referred to this literature.

The efforts to reduce NO\textsubscript{x} emissions were less successful (see section 4), as can be seen from the trend of NO\textsubscript{2} concentrations in Figure 9. From 1981 to 2000, there is a decrease of less than 40% in the urban background and more or less a stagnation afterwards. Apparently, the strong decrease of NO\textsubscript{x} emissions from power plants by the denitrification (see section 3.2) was partly counterbalanced by the enhanced traffic emissions (see section 4). Thus the additional NO\textsubscript{2} increment at traffic exposed stations remained high. NO concentrations reacted more sensitive to the decreasing NO\textsubscript{x} emissions than the NO\textsubscript{2} levels, giving rise to a gradual increase of the NO\textsubscript{2}/NO ratio over time. This shift is at least partly due to ozone chemistry and will be more thoroughly discussed in Part III.

From 1990 onwards, a slight increase of ozone concentrations at rural background sites was observed (Figure 10). This corresponds to slightly increasing or stagnant ozone background concentrations observed at many sites in Europe [91]. On the other hand, there was a distinct decrease of ozone peak concentrations during summer smog episodes, represented by the number of days per year exceeding the information threshold (180 µg/m\textsuperscript{3}, hourly mean) and the alarm threshold (240 µg/m\textsuperscript{3}, hourly mean) of the European Ozone directive [92] as shown in Figure 10b. The differences in weather conditions in the summers in Northwestern Germany are reflected by the strong variations of the ozone peak levels from year to year (see for example the high exceedances in the hot summer 2003).

Whereas annual ozone background levels in Europe can hardly be influenced by local or regional abatement measures alone, peak ozone concentrations can effectively be reduced by regional or national reductions of precursor emissions. The abatement measures for VOC from in-
dustrial sources (see sections 3.2 and 4), from the storage and distribution of fuels (see section 3.3) and from traffic by introducing the three way catalyst (see Part III) first lowered the VOC concentrations in ambient air and subsequently also ozone peak concentrations.

By the year 2000, there were almost no more exceedances of the German air quality standards enshrined in the “Technical Instructions” from 1986 [7] except for the deposition of some heavy metals in the vicinity of the heavy industry. The job to clean up the air in the Ruhr district seemed to be more or less accomplished. However, important new developments such as the assessment of air quality by the World Health Organization (WHO) had not been completely realized. This will be described in the upcoming Part III.

6 Conclusions

The second period of air pollution control accomplished a net decrease of the industrial emissions of all tackled pollutants, often by an order of magnitude or more.

For some pollutants (NOx, to a lesser extent VOC) this decrease was partly counterbalanced by high emissions from other sources such as traffic or the evaporation of solvents from products.

Ambient air quality improved distinctively as expected with the decreasing emissions, particularly for SO2, dust and toxic components of suspended particulates, to a lesser extent for NO2 and ozone.

European legislation was not yet a major driver on the way to clean air. German legislation often played a pioneering role. However, around the end of the nineties, important new developments such as the assessment of air quality by the WHO were not realized fully.

The net improvement of air quality was caused by the following factors:

- the creation of a comprehensive basis of federal laws and ordinances, setting ambitious emission limits, based on the precautionary principle,
- an efficient implementation of new legislation by the competent authorities, particularly by dedicated abatement programs,
- a fast development of flue gas cleaning technology and assessment methods,
- a trustful cooperation at the federal and state level, partly compensating for gaps in the federal legislation,
- structural change and a concentration process of the heavy industry,
- reduction of long range transport from Eastern Germany and Eastern Europe,
- an effective monitoring of air quality, serving as benchmark for the success of abatement measures.

In a nutshell, it can be stated that the improvement is an example of an efficient “command and control” approach, fostered by a fast development of best available techniques, assessment methods and favorable economic conditions. Furthermore, the reunification of Germany and the changes of the political and economic situation in Eastern Europe since 1989 added to the success in air pollution abatement.