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

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**Abstract** Part III of the short history of air pollution control covers the period from 2000 up to present. The requirements of the European legislation on clean air and on adjacent fields such as nature protection are outlined. These were the major drivers and basis of important amendments of the national law during the last decade. The continued modernization of industrial facilities and clean air plans addressing also the impacts from traffic led to decreasing emissions (with the exception of ammonia) and a further improvement of air quality. However, limit values for  $NO_2$  and to a lesser extent for  $PM_{10}$  are still exceeded. Another problem unsolved so far is the high input of nitrogen compounds into ecosystems, leading to eutrophication.

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

Der dritte Teil der kurzen Geschichte der Luftreinhaltung beschreibt den Zeitraum von 2000 bis heute. Die Anforderungen der Europäischen Gesetzgebung zur Luftreinhaltung und auf benachbarten Feldern wie dem Naturschutz werden umrissen. Diese waren die treibenden Kräfte im letzten Jahrzehnt und die Grundlage wichtiger Ergänzungen des nationalen Rechts. Die fortgesetzte Modernisierung der Industrieanlagen ebenso wie neue Luftreinhaltepläne, die auch die Auswirkungen des Verkehrs behandeln, führten zu sinkenden Emissionen (mit Ausnahme von Ammoniak) und einer weiteren Verbesserung der Luftqualität. Jedoch werden nach wie vor Grenzwerte für  $NO_2$  und in geringerem Maße auch für Feinstaub ( $PM_{10}$ ) überschritten. Ein weiteres ungelöstes Problem sind die hohen Einträge von Stickstoffverbindungen in Ökosysteme, die zu Eutrophierung führen.

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### 1 Introduction

At the beginning of the present millenium, pollution levels – in particular dustfall, suspended particulates and  $SO_2$  – had been reduced to such an extent that visible air pollution no longer occurred (see Parts I [1] and II [2]). The blue skies over the Ruhr district were regained. Were blue skies also tantamount to clean air? Taking the current German ambient air quality standards as benchmark, enshrined in the "Technical Instructions on Air Quality Control" from 1986 [3], this seemed to be the case (Table 1). Except for limited exceedances of heavy metal depositions in the vicinity of the heavy industry installations, air quality standards were met, often markedly.

However, the current German air quality standards did not consider properly recent international research findings on the effects of air pollutants on human health, as expressed in the air quality guidelines of the World Health Organization (WHO) [5]. Particularly the pioneering US studies from Dockery et al. [9] and Pope et al. [10], demonstrating effects of fine particles on mortality well below current ambient air concentrations, gave rise to a more critical evaluation of the effects of fine particles (PM $_{10}$  and PM $_{2.5}$ ) on human health. Taking the WHO air quality criteria as benchmark, it became clear that clean air for everyone had not yet been achieved (Table 1).

The WHO air quality guidelines served as blueprint for the European limit values, so that additional efforts became necessary to go beyond the "blue skies".

Therefore a revived struggle for clean air occured, predominantly prompted by new EC regulations. This recent development is presented in Part III.

Table 1. Highest pollution burden measured in the year 2000 at stations in North Rhine-Westphalia, in comparison to air quality standards then in force.

Pollutant	German standards [3]	Highest pollution load <sup>c)</sup> [4]	WHO air quality guidelines	EC limit values [6 to 8]
	in µg/m³	in μg/m³	[5] in μg/m³	in µg/m³
TSP	150 (ann) <sup>a)</sup>	72	_	_
	300 (98%)	279	_	_
PM <sub>10</sub>	_	41	risk estimate	40 (ann) <sup>a)</sup>
	_	75 <sup>b)</sup>		35 <sup>b)</sup>
Pb in TSP	2.0 (ann) <sup>a)</sup>	0.6	0.5	0.5
Cd in TSP	0.04 (ann) <sup>a)</sup>	0.033	0.0005	0.0005 <sup>e)</sup>
CO	10,000 (ann) <sup>a)</sup>	900	10,000 (8 hour)	10,000 (8 hour)
SO <sub>2</sub>	140 (ann) <sup>a)</sup>	25	50 (ann) <sup>a)</sup>	_
_	400 (98%)	125	_	125 (24 h)
NO <sub>2</sub>	80 (ann) <sup>a)</sup>	46	40 (ann) <sup>a)</sup>	40 (ann) <sup>a)</sup>
_	200 (98%)	92	200 (1 hour)	200 (24 h) <sup>d)</sup>

a) annual mean; b) number of exceedances per year of a daily mean of 50 μg/m³; c) concentration at the monitoring station with the highest pollution load;

d) 18 exceedances per year allowed; e) target value

Table 2. The European air quality directives since 1996.

EC directive	Important regulations
Framework directive	List of priority pollutants, timetable for daughter directives, assessment of air quality on the whole
96/62/EC [12]	territory, action plans and air quality plans in the case of exceedances of the limit values, content of the plans
1st daughter directive	Limit values for PM <sub>10</sub> , NO <sub>2</sub> , SO <sub>2</sub> , Pb to be met immediately (SO <sub>2</sub> ) or in the future (e.g. for PM <sub>10</sub> :
1999/30/EC [8]	2005); alert thresholds for SO <sub>2</sub> and NO <sub>2</sub> , air quality assessment (monitoring, modelling), quality
	assurance, information of the public
2 <sup>nd</sup> daughter directive	Limit values for CO and benzene (to be met immediately and 2010, respectively) assessment, quality
2000/69/EC [9]	assurance, information as in 1 <sup>st</sup> directive
3 <sup>rd</sup> daughter directive	Long term air quality objectives and target values for O <sub>3</sub> , information and alert thresholds,
2002/3/EC [13]	assessment, information of the public
4 <sup>th</sup> daughter directive	Target values for As-, Cd-, Ni-compounds and PAH in PM <sub>10</sub>
2004/107/EC [10]	
Ambient Air Quality directive	Merging Framework-, 1st, 2nd, 3rd daughter directive, limit values for PM <sub>2.5</sub> , time extensions to meet
2008/50/EC [14]	limit values until 2011 (PM <sub>10</sub> ) and NO <sub>2</sub> , benzene (2015) at the latest

# 2 Updating the legal instruments

#### 2.1 European level

The fifth EC action program for the environment [11] had adopted the current WHO air quality guidelines as benchmark for clean air to be reached in Europe and as basis for the setting of air quality standards.

An important milestone on the long way to clean air in Europe was set in 1996 and in subsequent years by the generic framework directive on ambient air quality assessment and management [12] and by four daughter directives dealing with specific pollutants [8 to 10; 13] (Table 2). The daughter directives were based on current science, described in position papers by experts of the Member States [15]. Of particular importance was the requirement to set up action plans (actions to be taken in the short term) and air quality plans (actions to meet the limit values at target dates) with abatement measures whereever limit values were exceeded

Limit values applied, and still apply, to the whole territory of the Member States, including hot spots of the pollution burden, where exposure of the public occurred. For Germany this requirement implied a change in the monitoring strategy. The approach to measure pollution levels which are representative for a certain area was substituted by an exposure oriented approach, assessing the air quality in typical microenvironments such as the urban background, near busy streets or in the neighbourhood of industrial facilities. The framework directive [12] and the  $1^{\rm st}$  to  $3^{\rm rd}$  daughter directives [8; 9; 13] were merged and updated in 2008 [14; 16] (Table 2). Also requirements for the particle size fraction  ${\rm PM}_{2.5}$  were added, including the requirement to reduce the exposure of the general population.

The regulations on ambient air quality can be regarded as the first strand of the European clean air strategy (CAFE, [17]). The second strand consists of source and product related legislation. Examples in respect to industrial sources are the directive 2000/76/EC on the incineration of waste [18], the directive 2001/80/EC on large combustion plants [19], and in particular the overarching directive 96/61/EC concerning the integrated pollution prevention and control (IPPC) [20] which was updated in 2008 [21] and 2010 [22]. The last update merged the IPPC directive with the regulations for specific industrial facilities mentioned above.

The IPPC directive [20] introduced the principle that the permit for industrial facilities and their inspections had to cover all environmental media such as air, water or soil. Requirements for large industrial installations (about 50,000 in the EU) had to be based on reference documents describing the "best available techniques" (BREFS) established by European working groups involving experts from industry, from NGO and the government. The permits had to be updated according to the technical progress. At first, the BREFS served as benchmarks for the competent authorities. By the revision in 2010 [22], the range of emission limits laid down in conclusions derived from the BREFS became legally binding under certain conditions.

The source related regulations shall be complemented by a directive on medium combustion plants. A proposal from the Commission has recently been published [23].

Starting as early as 1970, the European Union had established regulations for the exhaust emissions of vehicles for pollutants such as  $\mathrm{NO}_x$ , VOC and particulates. An overview of the early regulations is presented in [24]. Since 1992, the three way catalyst for cars with spark ignition became compulsory (EURO1, [25; 26]). In the following the exhaust regulations for passenger cars and light duty vehicles (EURO1-6) and for heavy duty vehicles (EURO I-VI) have been updated, lowering the emissions in six stages. An overview is presented in Figures 1 and 2 [27]. The emission limits apply to the type approval of vehicles and refer to fixed test cycles (currently for the new european driving cycle, NEDC [28]).

The third strand of the European clean air policy sets limits for national emissions of certain pollutants (ceilings) specific for each Member State and was introduced in 2001 (NEC directive) [29], with a view to trigger national emission reduction plans and to reduce long range transport between the Member States. A similar intention is pursued by the Gothenborg protocol within the framework of the United Nations Economic Commission for Europe (UNECE) [30]. National emissions must be lowered in order to reduce pollutant input to ecosystems, ultimately leading to acidification, eutrophication and to photochemical air pollution. The emission ceilings set by the UNECE have recently been revised [31], and a proposal of the European Commission to revise the NEC directive is currently under discussion [32]. Table 3 summarizes the ceilings for Germany. They approximately correspond to the EC average or are less stringent (SO<sub>2</sub>).

In addition to the Clean Air Programme [17] important legislation in neighbouring fields such as nature conservation has to be mentioned. The FFH directive [33] in combination with the directive 2009/14/EC [34] for the protection of birds inter alia require the Member States to install a network of specially protected areas (Natura 2000). Projects such as the construction of industrial facilities or major roads have to be scrutinized for their potentially noxious impacts on these sites, such as acidification or eutrophication. Significant impacts which can damage the natural function are to be avoided.

#### 2.2 National level

The European regulations described above made several updates of the national legislation necessary. The Federal Immission Control Act [35] was amended several times, inter alia to transpose the European directives mentioned in section 2.1 into national law. An overwiew is given in [36]. Also the amendments of important ordinances such as the 10<sup>th</sup> on the quality of fuels [37], the 13th on large combustion plants [38] or the 17th on incinerators [39] were initiated through new European legislation. The 39th ordinance from 2010 [40; 16] implemented the directive 2008/50/EC [14; 41] and the NEC directive [29]. An innovative national element was the 35th ordinance on the labeling of vehicles according to their emissions [42] which is a legal prerequisite for setting up low emission zones.

Also the amendment of the "Technical Instructions on Air Quality Control" from 2002 [45] adopting the air quality standards of the 1<sup>st</sup> and 2<sup>nd</sup> daughter directive [8; 9] for the permit of industrial facilities was partly due to European legislation. In addition, stricter emission standards for numerous industrial installations were set, according to the progress in best available technologies. Existing facilities had to comply with these stricter emission limits by certain deadlines, generally by October 30<sup>th</sup> 2007 [43].

The EC directives on nature protection and conservation [33; 54] gave rise to amendments of the Federal Nature Conservation Act [44], in particular of articles 31 to 34 (Natura 2000 network, protection, impacts of projects). In the framework of industrial permits, projects (also the summation of several projects) have to be examined in respect to their impacts on the Natura 2000 network (FFH assessment). A European Court ruling from May 2011 [45] made clear that not only af-

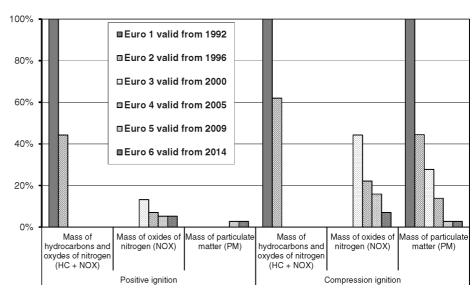


Figure 1. European exhaust regulations for passenger cars and light duty vehicles since 1992 [27].

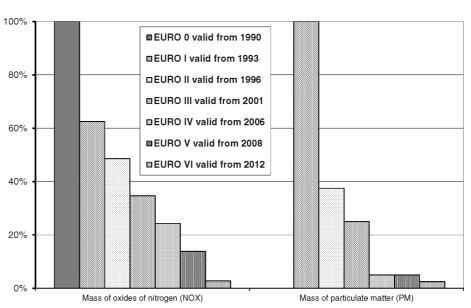


Figure 2. European exhaust regulations for heavy duty vehicles since 1990 [27].

Table 3. National emission ceilings for Germany (NEC), Kt/a.

Pollutant	2001/81/EC, ceilings for 2010 [29]	UNECE, 2005-2020 [30]	COM proposal, 2005-2030 [31]
SO <sub>2</sub>	520	-21% (410)	-53% (244)
NO <sub>x</sub>	1,051	-39% (901)	-69% (458)
VOC	995	-13% (995)	-43% (652)
NH <sub>3</sub>	550	-5% (545)	-39% (350)
PM <sub>2.5</sub>	_	-26% (90)	-43% (69)
CH <sub>4</sub>	_	_	-39%

fected citizens, but also NGO's are entitled to apply to national courts regarding FFH assessments.

#### 3 Abatement measures in North Rhine-Westphalia

In contrast to 1986 (see Part II, section 3.2), the amendment of the "Technical Instructions on Air Quality Control" in 2002 [43] did not give rise to a special programme for the update of industrial facilities. Instead, this modernization was part of

Table 4. Two examples of air quality plans

Case	Duisburg-Bruckhausen	Düsseldorf-Corneliusstraße
Exceedances giving rise to first plan (year)	PM <sub>10</sub> , 128 d > 50 μg/m³ (2002) PM <sub>10</sub> , 46 μg/m³ (annual, 2002)	PM <sub>10</sub> , 108 d > 50 μg/m³ (2003) PM <sub>10</sub> , 45 μg/m³ (annual, 2003) NO <sub>2</sub> , 62 μg/m³ (annual, 2003)
First plan	Luftreinhalteplan, 2004	Luftreinhalteplan, 2004
Current plan (several updates in between)	Luftreinhalteplan Ruhrgebiet-West, 2011 [47]	Luftreinhalteplan Düsseldorf, 2013 [50]
Source apportionment in 2004 [46] (percentage of local pollution burden, %)	Local industry 39 Road traffic 6 Regional background 51	NO <sub>2</sub> : Industry 1 Road traffic 53 Regional background 36 PM <sub>10</sub> : Industry < 1 Road traffic 27 Regional background 62
Abatement measures (selection)	Replacement of old cokery (2003), upgrade of electrostatic filters, encasement and filtering of diffusive sources (2004, 2005); new filters for blast furnaces (2004); low emission zone (LEZ) (2008); replacement of a blast furnace by a new one (2007); reduction of diffusive emissions from stockpiles (2007); fabric filter for sintering plant (2010)	Routing concept for HDT (2005); traffic bans for HDT > 2,8 t (2005); LNG-buses (2005); improvement of traffic flow (2005); low emission zone (LEZ) (2009); enlargement of LEZ (2013); upgrade of municipal vehicles (LNG, EURO5 or better); optimization of public transport (2010); traffic management system (2010); promotion of bicycles and of electromobility (2012)

the routine enforcement by the competent authorities. Nevertheless, a further reduction of industrial emissions could be achieved.

Due to frequent exceedances of the limit values for  $PM_{10}, NO_2$  and in two cases for benzene, numerous air quality and action plans had to be established. Currently, 34 air quality plans are in force and four additional plans are in preparation. All exceedances of the  $NO_2$  limit value occur at traffic hot spots, predominantly in busy street canyons. Exceedances for  $PM_{10}$  are both measured near industrial facilities with strong diffusive dust emissions (steel mills, harbour regions, quarries, lignite pits) and near traffic hot spots, whereas the exceedances for benzene were caused by industrial emissions.

The information to be included in the air quality plans is laid down in Annex XV of the directive on ambient air quality [14] and comprises, inter alia, the localization and assessment of the pollution, a source apportionment based on the emissions and on model calculations quantifying the contribution of all known sources (e.g. traffic urban background, long range transport), and details of the abatement measures including a forecast of their impact.

Depending on the source apportionment, air quality plans may refer to the local scale with areas of several square kilometers near a specific industrial source or to a short length of a busy street canyon in a small or medium sized city [46]. They may also cover a whole region such as the western or eastern part of the Ruhr district (regional plans) [47]. Like-

wise, the abatement measures may be focussed on a particular industrial installation [46] a local traffic situation [46] or on the traffic network of several adjacent cities [47]. In particular for exceedances caused by traffic, there is a shift from the local scale to regional air quality plans [47 to 49]. Model calculations for conurbations have shown that high pollution loads not only occur at isolated hot spots, but affect larger parts of the traffic network. Consequently, exceedance situations which were originally dealt with by local air quality or action plans [46] are now approached by regional air quality plans [47 to 49]. Table 4 presents two examples, and Figures 3 and 4 show the trend of PM<sub>10</sub> concentrations at the two corresponding monitoring stations. The trends demonstrate the net improvement of the air quality brought

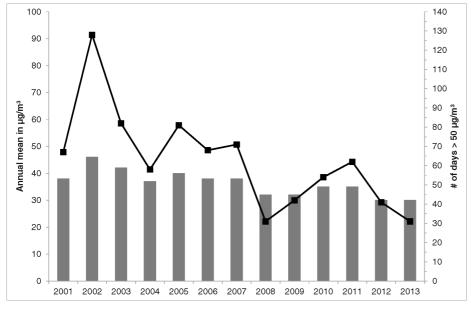


Figure 3. Trend of the  $PM_{10}$  concentrations (annual means,  $\mu g/m^3$ ) and number of days in exceedance of a daily mean of 50  $\mu g/m^3$  at the monitoring station Duisburg-Bruckhausen near a steel mill.

forward by the abatement measures listed in Table 4 and by technical developments due to the European exhaust regulations. In the case of the traffic hot spot Düsseldorf-Corneliusstraße, carbon concentrations (elemental and organic carbon) could significantly be reduced as well.

One regional measure to reduce the air pollution caused by traffic has already been mentioned, namely low emission zones (LEZ). LEZ restrict admission to certain zones (for example city centers) for vehicles with higher exhaust emissions, for example cars not meeting the requirements of EURO4 or EURO3 (see Figure 1). An enforcement is made possible by the labeling of vehicles according to their exhaust emissions [42]. At present, 13 LEZ are in force in North Rhine-Westphalia. By July 2014, only vehicles meeting the EURO4 (passenger cars) or EURO IV (trucks) standards are entitled to circulate within these zones. Besides a traffic ban for vehicles with higher emissions in certain areas, the LEZ encourage the modernization of the fleet. The LEZ established in the Ruhr district covering an area of 868 km<sup>2</sup> is one of the largest zones in Europe. This regional measure affects not only traffic hot spots, but comprises the whole traffic network.

The effectiveness of LEZ is debated. Whereas several authors [51] have found reductions of the pollution burden at traffic hot spots in the range of 2.1 to 2.4  $\mu g/m^5$  (PM $_{10}$ ) and 1.2 to 3.7  $\mu g/m^5$  (NO $_2$ ) after correcting for the general trend of air pollution, one research group came to the conclusion that the effects on PM $_{10}$  concentrations within LEZ are small [52]. However, this group did not only consider traffic exposed stations, but included also urban background stations in their analysis, where the additional pollution increment by local traffic and consequently also the reductions are small.

As an effect of LEZ, a faster modernization of the fleet was found within the Ruhr district opposed to North Rhine-Westphalia as a whole [51].

# 4 Development of the emissions

Data on the emissions of important air pollutants ( $PM_{10}$ ,  $NO_x$ ,  $NH_5$ ) from different source categories are presented in Figures 5 to 7 for the years 2000 to 2012.  $PM_{10}$  and  $NO_x$  data are from the North Rhine-Westphalian emission inventory [53]. Industrial emissions were calculated every  $4^{th}$  year from data collected within the framework of the  $11^{th}$  ordinance [54] to the Federal Immission Control Act, where-

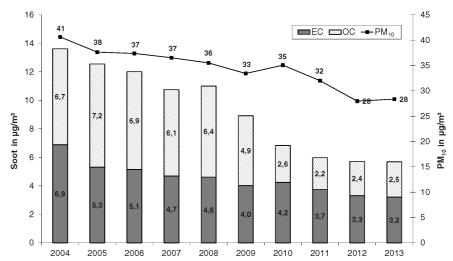


Figure 4. Trend of  $PM_{10}$ , elemental carbon (EC) and organic carbon (OC), annual means ( $\mu g/m^3$ ) at the monitoring station Düsseldorf-Corneliusstraße, placed in a busy street canyon.

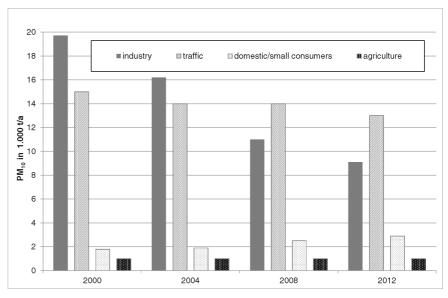


Figure 5. Annual  $PM_{10}$  emissions (1,000 t/a) in North Rhine-Westphalia from different source categories in the years 2000 to 2012.

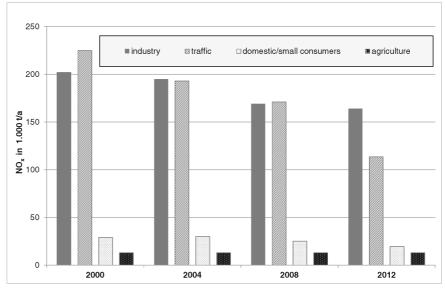


Figure 6. Annual  $NO_x$  emissions (1,000 t/a) in North Rhine-Westphalia from different source categories in the years 2000 to 2012.

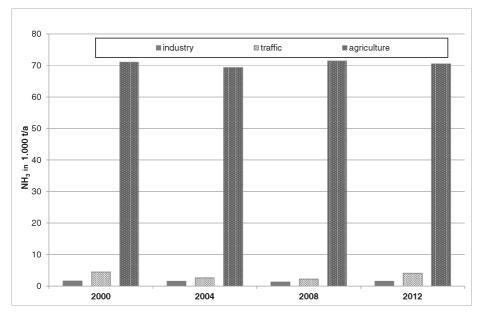


Figure 7. Annual  $NH_3$  emissions (1000 t/a) in North Rhine-Westphalia from different source categories in the years 2000 to 2012.

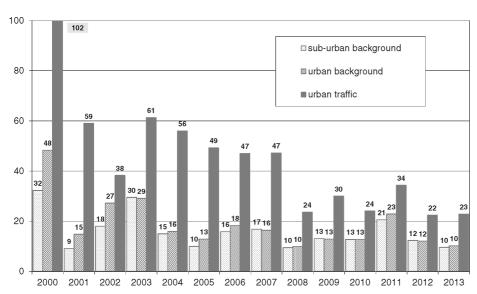


Figure 8. Trend of the number of days per station in exceedance of a  $PM_{10}$  daily means of 50  $\mu g/m^3$  at traffic exposed sites (average over all sites) and in the urban background.

as emissions from traffic and domestic sources and small consumers were calculated based on activity patterns and emission factors.  $NH_3$  emissions from agriculture were taken from emission models run for Germany, broken down to North Rhine-Westphalia [55].

A considerable decline of industrial  $PM_{10}$  emissions by about 50% can be read from Figure 5, whereas  $PM_{10}$  emissions from traffic decreased only slightly. The decrease of the exhaust emissions is partly masked by constant emissions from dust resuspension and abrasion, which depend on the traffic volume. Please note that  $PM_{10}$  emissions from the domestic sector are rising, due to increased burning of biomass.

With regard to  $\mathrm{NO_x}$  emissions (Figure 6), there is a further slight decrease in industrial emissions by about 19% since 2000 and a net decrease in traffic emissions, particularly from 2008 to 2012. Figure 7 shows that  $\mathrm{NH_5}$  emissions from agriculture remained constant. The contributions from the other source categories were small. When it comes to nitrogen deposition,  $\mathrm{NO_x}$  emissions from industry and  $\mathrm{NH_5}$  emissions

sions from agriculture are of comparable size, both corrected for their different molecular weights.

# 5 Trend of the air quality

It has already been mentioned that the European legislation on air quality [8 to 14] gave rise to a change in the assessment strategy from an area representative to an exposure oriented approach. Whereas in the year 2001 seven continuous stations were operating near traffic hot spots in the monitoring network of North Rhine-Westphalia, this number has risen to 23 traffic exposed stations in 2013.

For NO<sub>2</sub>, the monitoring stations are complemented by a dense network of passive samplers, of which about 75 are near busy streets.

The second column of air quality assessment, the modelling of air quality, has become more and more important within the last decade, fostered by the improved quality of the models and emission inventories and also by the requirement of the European legislation to assess air quality on the whole territory of the Member States [14]. Modelling is indispensable for source apportionment [56], for obtaining an overall picture of the air quality, for measurement planning and for assessing the effects of abatement measures. The placing of new monitoring stations was guided by a systematic screening of traffic hot spots by modelling [57], and the delimitation of the low emission zones was based on modelling the impacts of the whole traffic network in the Ruhr region on air quality [58].

The results from the monitoring net-

work show clearly that the limit values of  $PM_{10}$  and  $NO_2$  were exclusively exceeded at hot spots (traffic or industrial) and not at urban or rural background stations. The stations have to be sorted by type, separately evaluated and normalized by the number of stations to derive a meaningful trend. The normalization was performed and the results are presented in  $\pmb{\text{Figure 8}}.$ 

A distinct decrease of the number of days per year exceeding a PM $_{10}$  daily average concentration of 50  $\mu g/m^{5}$  (days in exceedance) at traffic exposed stations and in the urban background is apparent. The considerable interannual fluctuation is related to different meteorological dispersion conditions. It can be concluded that the EC exhaust regulation, in particular the introduction of particulate filters, in combination with the additional abatement measures discussed in section 3 was effective.

This is unfortunately not the case for  $NO_2$  concentrations at traffic exposed sites which were more or less stagnant within the last decade (see Figure 9 in Part II [2]). It seems that at

least the maximum of the  $NO_2$  burden in the years 2008 and 2009 has now been passed. Since the year 2010, the  $NO_2$ 

levels are slowly declining. However, even the average NO2 concentrations at traffic exposed sites still exceed the limit value of  $40 \,\mu g/m^5$  [14] which was to be met by 2010. Recent research revealed the reasons for the failure of the European exhaust regulations which should have led to a distinct reduction in NOx emissions (see Figures 1 and 2). First, the NO to NO<sub>2</sub> ratio has changed over time which can be seen from a long time series at the traffic exposed station Essen-Ost in Figure 9 and also from Figure 9 in Part II [2]. Initial significant reductions in NOx emissions led almost exclusively to a reduction of NO levels, the NO<sub>2</sub> concentrations remaining more or less constant. The main reason for this change of the NO to NO<sub>2</sub> ratio are the oxidation catalysts of vehicles with compression ignition, raising the NO<sub>2</sub> share of the exhaust emissions from about 5% (EURO1, EURO I-III) up to 35 to 50% (EURO4, EURO5) and 10 to 15% (EURO IV, EURO V), respectively [59]. Only catalytic reduction (SCRT, EURO VI) will lower this ratio again. The change of the NO to NO2 ratio observed in street canyons [60] is enhanced by ozone chemistry (with decreasing NO concentrations, more ozone is available for the rapid oxidation of NO to NO2 near the source) [60] and by the rising share of diesel vehicles in the fleet.

A further reason for the stagnant NO<sub>2</sub> levels in the last decade is the large

discrepancy between real world  $\mathrm{NO_x}$  emissions and the emissions measured during the European type approval [61; 62]. The test cycle NEDC with only mild accelerations and low engine loads strongly underestimates the  $\mathrm{NO_x}$  emissions of diesel cars under urban congested driving conditions [61]. Up to five times higher emissions compared to the NEDC have been found [63], and  $\mathrm{NO_x}$  emissions from EURO5 diesel passenger cars even slightly exceed those from EURO1 vehicles under real world driving conditions. Not before EURO6 and EURO VI vehicles will reach a significant percentage of the fleet,  $\mathrm{NO_2}$  concentrations at traffic hot spots will drop decisively. In addition, the NEDC for passenger cars will be replaced in 2017 by a more realistic type approval [63], whereas the type approval for heavy duty vehicles has already been improved by the EURO VI stage [62].

Another persisting environmental problem is the high input of nitrogen compounds into ecosystems, which frequently exceed critical loads and thus lead to eutrophication. In Germany, about 78% of the area of ecosystems are affected [64], and in the European Union 62% [63]. The high deposition of nitrogen compounds is caused by the high  $NH_3$  emissions

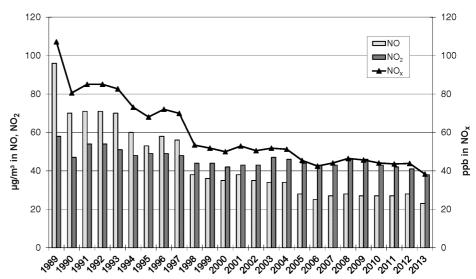


Figure 9. Trend of NO,  $NO_2$  concentrations (annual means,  $\mu g/m^3$ ) and  $NO_x$  concentrations (ppb) at the traffic exposed monitoring station Essen-Ost.

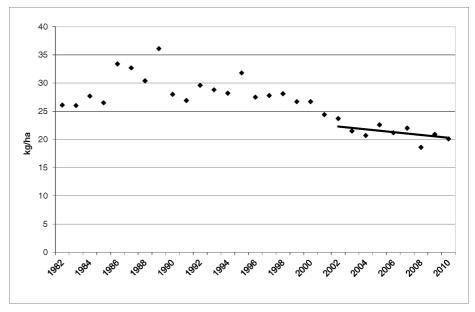


Figure 10. Measured nitrogen deposition (kg per hectar and year) into forests in North Rhine-Westphalia [65].

from agriculture (about 90% of the  $NH_{\rm 5}$  emissions) and to the  $NO_x$  emissions from traffic and industry.

Figure 10 shows the trend of the nitrogen deposition into the forests in North Rhine-Westphalia [65], measured since 1983. Whereas a decrease during the first two decades was observed, reflecting the decrease of  $\mathrm{NO}_x$  emissions (see Part II, section 3), the trend has nearly levelled out in recent years due to the persisting high  $\mathrm{NH}_5$  emissions from agriculture. Inter alia, this complicates the FFH assessment within the framework of permitting new installations, as in many areas of the Natura 2000 network, the critical loads for the nitrogen deposition are exceeded.

#### 6 Outlook

Although the blue skies over the Ruhr district have been regained and notwithstanding the important improvement of air quality brought forward by air quality control and structural change, clean air for every citizien has not yet been achieved. In particular, two big problems persist:

At traffic hot spots, there are still several exceedances for the limit value for PM<sub>10</sub> (daily means) and widespread exceedances for the limit value for NO<sub>2</sub> (annual means). Apart from the implications on human health there is a pending risk of infringment procedures against Germany. For NO<sub>2</sub>, the reasons for the persisting breaches of the limit values at traffic hot spots have been discussed in section 5. It will probably take until 2020 and beyond, until a significant share of the fleet will have been replaced by vehicles meeting the EURO6 and VI standards which hopefully will reduce the  $NO_2$  levels significantly. When it comes to  $PM_{10}$ , emissions from small stoves burning biomass in the domestic sector without any flue gas cleaning should not be overlooked, as the number of small stoves has soared in recent years [66] in parallel to rising energy prices. Whereas the contribution of biomass burning to average PM<sub>10</sub> levels is small [66], the impact on the number of exceedances of  $PM_{10}$  daily means is considerable, reaching up to 20 to 50% of the exceedances during cold winters with frequent inversions [66]. Notwithstanding the general synergies between climate change policies and air pollution control, this is one example of an important trade-off. Action should be taken not to risk one important achievement of the first decades of air pollution control, the structural change of domestic heating away from burning solid fuels in small stoves.

The problem of still elevated levels of fine particles and  $\mathrm{NO}_2$  is exacerbated by recent research which has strengthened the evidence for noxious effects of these compounds on human health. The WHO has collected this new evidence within two recent reviews (REVIHAAP [67] and HRAPIE [68]), and the IARC has classified particles in ambient air as human carcinogen [69]. Consequently, a further reduction of air pollution is called for, and it is highly probable that the European Commission will come forward with proposals for lower limit values, once it comes to a revision of the air quality directive 2008/50/EC [14] in 2020. The "distance to target" will probably increase again. Within this context, also the effects by other particle fractions such as ultrafine parti-

cles on human health will have to be evaluated including a decision whether the particle mass as principle metric for standard setting will have to be complemented by other metrics such as particle numbers.

Another unsolved problem is the high deposition of biologically available nitrogen (NO<sub>x</sub> and NH<sub>y</sub>) into protected areas, exceeding the critical loads in many sensitive ecosystems [64]. Whereas NO<sub>x</sub> emissions are decreasing, NH<sub>3</sub> emissions predominantly from agriculture are constant. This holds true not only for North Rhine-Westphalia, but for Germany as well [64] and for great parts of the European Union [63]. It is evident that after industry and traffic have delivered more or less their share in air pollution abatement, it is now the turn of the agricultural sector to carry on. Particularly the industrial lifestock production with their secondary effects such as manure management has to be addressed, and European as well as national action plans for nitrogen management are called for. The excessive nitrogen input into ecosystems has not only become a problem for ecosystems but also hinders the authorization of industrial facilities. Procedures are under development to deal with this problem in a reasonable way [70].

In addition to nitrogen deposition, emissions of bioaerosols are an upcoming topic. Effects on the respiratory system of people living near several big stables have been described [71], and elevated emissions of endotoxins and other bioaerosols from stables have been measured [72; 73]. A guideline to assess the effects of bioaerosols on human health has recently been published [71]. This assessment is difficult, though, as no quantitative dose response relationships could hitherto be established. Nevertheless this open question will intensively be adressed by concerned citizens.

We may conclude that inspite of the long distance already covered on the road to clean air, the aim has not yet been reached completely.

Literature references are available from www.gefahrstoffe.de, Section Supplementary Material.