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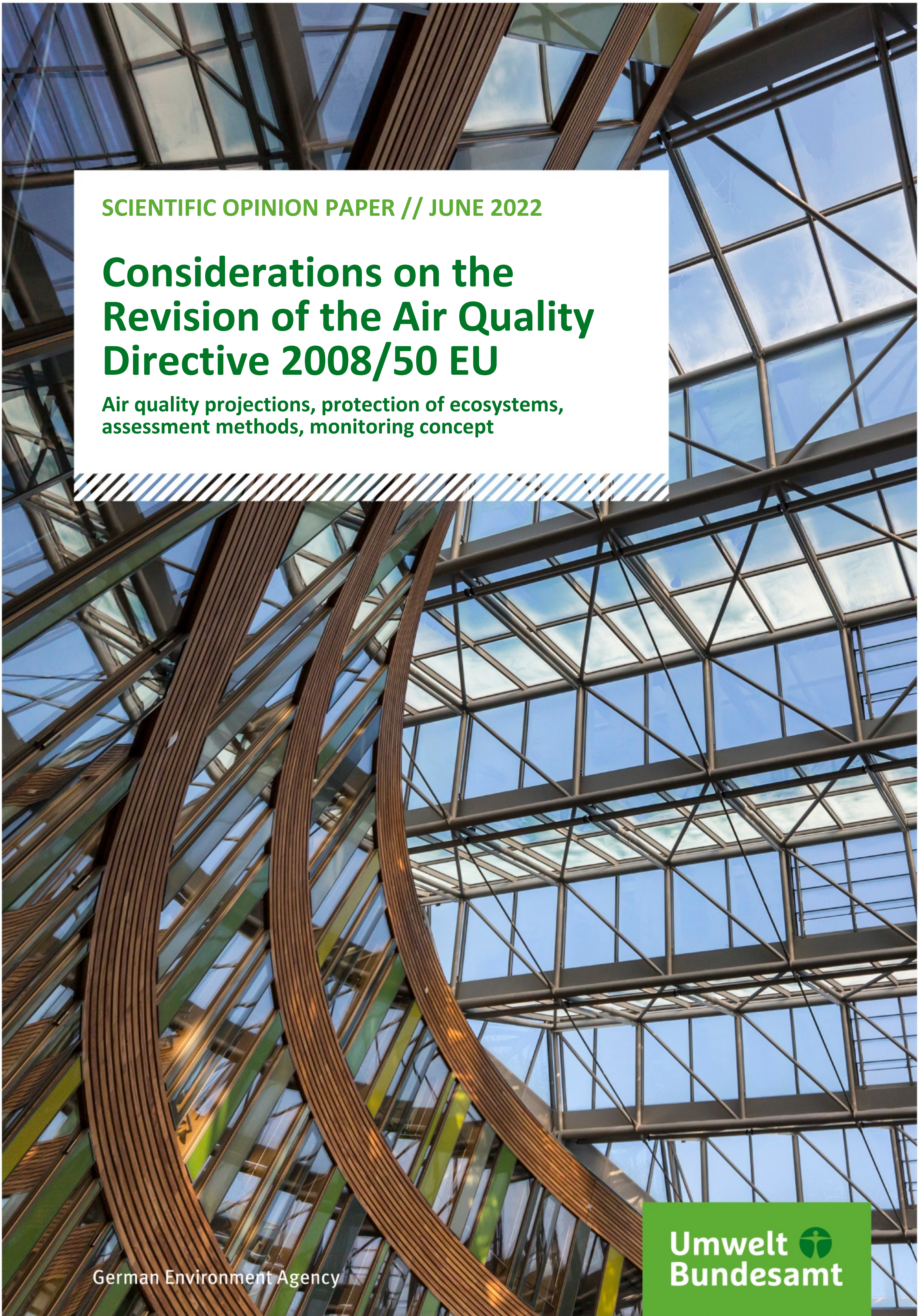
Air quality projections, protection of ecosystems, assessment methods, monitoring concept



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Considerations on the Revision of the Air Quality Directive 2008/50 EU

Air quality projections, protection of ecosystems, assessment methods, monitoring concept

by

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On behalf of the German Environment Agency

Executive Summary

The concept of EU Air Quality Legislation was laid down 25 years ago by agreeing on the Ambient Air Quality Framework Directive 96/62/EU. This regulation has been supported by four so called Daughter Directives which have been mainly compiled in the current Ambient Air Quality Directive (AAQD) 2008/50/EU without much substantial change.

In the past decades, the state of science has developed in many aspects: Automated monitoring methods have been improved, uncertainties of air quality modelling results have been reduced and the evidence on the risk to ecosystems by air pollutants has grown. Last but not least the **World Health Organisation (WHO) has issued new Air Quality Guidelines (AQG)** in 2021, piling up the evidence on adverse health effects resulting from exposure to air pollutants and thereby underlining the need for a revision of the current AAQD.

In 2021, the European Commission published the **Zero Pollution Action Plan** which proposes a long-term "zero pollution vision for 2050", which includes the goal of reducing air pollution, among others, to "levels no longer considered harmful to health and natural ecosystems."

Consequently, the European Commission has announced a **revision based on the current AAQD** focussing on three policy areas:

- Policy area 1: closer alignment of the EU air quality standards with scientific knowledge including the latest recommendations of the WHO
- Policy area 2: improving the air quality legislative framework, including provisions on penalties and public information
- Policy area 3: strengthening of air quality monitoring, modelling and plans

In preparation for the upcoming revision the German Environment Agency (UBA) from a scientific point of view has assessed the **possibility to meet the proposed WHO Guidelines Levels**, the **applicability of improved chemical transport models (CTM)** and **advanced monitoring methods for air quality assessment** as well as concepts for a **better protection of ecosystems from air pollutants**. In the following we propose criteria for binding and non-binding air quality standards as well as a monitoring strategy beyond compliance checking. All considerations All considerations are based on the German situation but may be extrapolated to many other European Member States (MS).

By implementing the National Air Pollution Control Programme as required by Directive (EU) 2016/2284 on the Reduction of National Emissions (NEC Directive), air quality in Germany will substantially improve until 2030. However, apart from PM_{10}, CO and SO_2 the AQG Levels proposed by WHO will not yet be attainable by cost-efficient measures at this date. From our projections for annual mean values at background locations we conclude that for **NO_2** WHO Interim Target 3 could be met as well as Interim Target 4 for **$PM_{2.5}$**. As a consequence, we recommend to better align the EU Air Quality Standards (AQS) with the latest WHO AQG by 2030 by tightening the current annual limit values to **20 µg/m³ for NO_2**, to **15 µg/m³ for PM_{10}** and to **10 µg/m³ for $PM_{2.5}$**. As it is very difficult to develop further national abatement strategies to substantially reduce the ozone levels we recommend a target value preferably stricter than WHO Interim Target 1, i.e. 100 µg/m³, as appropriate for **ozone**.

While these recommendations are based on available abatement measures, we consider it necessary to **continuously improve air quality beyond 2030**, aiming to achieve the stricter WHO AQG levels by a step-by-step reduction of the proposed Air Quality Standards (AQS).

As a prerequisite for a better alignment of the EU AQS with the latest WHO AQG we consider a **modified governance structure** of high importance. The future AAQD should include provisions that ensure both coherence and transparency between different levels of governance, e. g. a provision requiring Union institutions and Member States to cooperate and take the necessary measures at Union and national level.

Not only human health but also ecosystems are endangered by current air pollution levels. Therefore, the AAQD should continue considering the **protection of ecosystems from ozone**. However, we propose that the POD concept (Phytotoxic Ozone Dose) should be used for the assessment instead of the AOT (Concentration Accumulated Over a Threshold) and an appropriate target value should be set. The AQS and the monitoring concept should be adapted accordingly. Additionally, we recommend the monitoring of ammonia to be incorporated into the revised AAQD, as it has significant adverse effects on biodiversity and ecosystems.

Air quality assessment should be improved by allowing for state-of-the-art monitoring methods and mandatory use of quality proven CTM. We also propose to make the use of **CTM mandatory in air quality planning**. However, we support the current concept of allowing only established reference methods to check compliance with limit values.

To understand the formation of secondary pollutants and to identify relevant emissions by source apportionment, we consider it necessary to **extend monitoring beyond compliance checking**. Harmonised observations at additional so-called **supersites** and for more substances would help to achieve this aim. In particular we consider it necessary to introduce mandatory monitoring for **Ultrafine Particles** and **Black Carbon**, thereby following the WHO, which has explicitly pointed out the risk to human health by these pollutants, underlining that additional data and health studies would be needed before an AQS could be recommended.

Content

1 Current and future air quality in Germany

In 1987 WHO for the first time issued AQG aiming at the protection of human health from air pollutants (WHO 1987). Since then the AQG have been updated frequently according to latest scientific evidence. The recent update in 2021 gives new guideline levels and interim targets for PM_{10}, $PM_{2.5}$, NO_2, Ozone, SO_2 and CO. For the first time WHO also recommends best practice examples for Ultrafine Particles (UFP) and Black Carbon/Elemental Carbon (BC/EC).

In order to identify the most harmful air pollutants we evaluated todays air quality in relation the new AQG. With a view to setting more ambitious AQS for 2030 we considered what improvements would be achieved by current and already planned upcoming legislation.

1.1 Air Quality in 2020

In Germany ambient air quality has substantially improved in the past decades. However, most of the new AQG Levels are only met in parts of the country. According to air quality monitoring under Directive 2008/50/EU the following situation in relation to AQG Levels and Interim Targets has been observed in 2020.

As can be seen from the Table 1 the AQG Levels are only met for CO at all sites and for SO_2 at nearly all sites. As for $PM_{2.5}$ Interim Target 3 and for PM_{10} Interim Target 4 are attained almost everywhere. For NO_2 only Interim Target 1 for the annual mean – corresponding to the current limit value - is met at most sites, while Interim Target 2 is exceeded at about 20 % of the sites. For ozone Interim Target 2 is exceeded almost everywhere.

Table 1: Exceedance of WHO AQG Levels and Interim Targets at German monitoring sites in 2020

Pollutant		Share of Stations in exceedance				
		Interim Target 1	Interim Target 2	Interim Target 3	Interim Target 4	AQG Level
$PM_{2.5}$	Annual mean	0% > 35 µg/m^3	0% > 25 µg/m^3	0% > 15 µg/m^3	14% > 10 µg/m^3	99% > 5 µg/m^3
	24 h[a] mean	0% > 75 µg/m^3	0% > 50 µg/m^3	2% > 37.5 µg/m^3	78% > 25 µg/m^3	99.5% > 15 µg/m^3
PM_{10}	Annual mean	0% > 70 µg/m^3	0% > 50 µg/m^3	0% > 30 µg/m^3	5% > 20 µg/m^3	36% > 15 µg/m^3
	24 h[a] mean	0% > 150 µg/m^3	0% > 100 µg/m^3	0% > 75 µg/m^3	7% > 50 µg/m^3	16% > 45 µg/m^3
O_3	Peak[b] season	3% > 100 µg/m^3	100% > 70 µg/m^3	no Interim Target	no Interim Target	100% > 60 µg/m^3
	8 h[a] mean	0.4% > 160 µg/m^3	95% > 120 µg/m^3	no Interim Target	no Interim Target	99.6% > 100 µg/m^3
NO_2	Annual mean	1% > 40 µg/m^3	22% > 30 µg/m^3	51% > 20 µg/m^3	no Interim Target	83% > 10 µg/m^3
	24 h[a] mean	0% > 120 µg/m^3	21% > 50 µg/m^3	no Interim Target	no Interim Target	76% > 25 µg/m^3
SO_2	24 h[a] mean	0% > 125 µg/m^3	2% > 50 µg/m^3	no Interim Target	no Interim Target	2% > 40 µg/m^3
CO	24 h[a] mean	0% > 7 mg/m^3	no Interim Target	no Interim Target	no Interim Target	0% > 4 mg/m^3

[a] 99th percentile

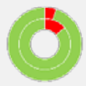
[b] Average of daily maximum 8-hour mean O_3 concentration in the six consecutive months with the highest six-month running-average O3 concentration (here: April-September)

1.2 Air Quality in 2030

To evaluate air quality in 2030, the impact of implementing the National Air Pollution Control Programme as required by the NEC Directive (EU) 2016/2284 was considered. The box below describes data and methods used to obtain the modelling results shown in Table 2.

The model results – applying to background stations only - show full compliance with at least Interim Target 3 for annual means of both, $PM_{2.5}$ and PM_{10}, and only minor exceedances of Interim Target 4 for PM_{10}. However, we expect exceedances of the AQG Level for $PM_{2.5}$ nearly everywhere. Modelling results for NO_2 show only few exceedances of Interim Target 3 but still obvious exceedances of the AQG Level. For ozone, Interim Target 1 will be met everywhere, but from Interim Target 2 onwards nearly all stations will be in exceedance.

Table 2: Exceedance of WHO AQG Levels and Interim Targets for annual means ($PM_{2.5}$, PM_{10} and NO_2) or Peak Season (O_3) in Germany in 2030 based on modelling results. For absolute values of AQG Levels and Interim Targets see table 1.

Pollutant	Share of grid cells with monitoring stations in 2020 with exceedances in 2030 (outer ring, red) and share of stations in exceedance in 2020 (inner ring, red)				
	Interim Target 1	Interim Target 2	Interim Target 3	Interim Target 4	AQG Level
$PM_{2.5}$					
PM_{10}					
O_3			No Interim Target	No Interim Target	
NO_2				No Interim Target	

Data and methods used for modelling air quality

Meteorological data:
Aiming for a conservative approach meteorological data from 2018 were used, a relatively warm and dry year with comparingly high PM-concentrations. The same meteorological conditions were assumed for 2030.

Emission data:
National emissions for 2018 were obtained from the submission of 2021[1]. For 2030, an updated version of the 'with additional measures' (WAM) scenario from the submission 2021 was used. The WAM scenario aims to be compliant with the emission reduction commitments (ERC) set up by the NEC Directive (EU) 2016/2284. These national emissions were gridded using the GRETA-tool (Gridding Emission Tool for ArcGIS) (Schneider et al. 2016).

Chemical Transport Model (CTM) and data assimilation:
Ambient concentrations were computed by using the regional CTM REM-Calgrid (Stern 2009, Stern et al. 2008, Nordmann et al. 2020) with a spatial resolution of about 7x8 km². Since the model results are typically biased, the model output was adjusted by applying an offset. The offset was

[1] Details of the submission can be found on https://cdr.eionet.europa.eu/de/eu/nec_revised/projected/envyaxjsa

calculated for 2018 using optimum interpolation as a data assimilation method with the software FLADIS (IVU Umwelt GmbH 2020) and measured data from 2018. The offset obtained for 2018 was also used to adjust the model output for 2030. From the modelled results for 2018 and 2030, concentrations for 2020 were calculated by linear interpolation for each cell. Afterwards, the relative concentration changes from 2020 to 2030 in the grid cells where observation sites are located were multiplied by measured data in 2020 and aggregated to obtain the annual mean concentrations for each cell in 2030 for different station types. The evaluation was confined to those grid cells where urban or rural background measurement stations were located in 2020.

2 Health relevant air quality standards

As announced by the EU Commission current air quality standards should be closer aligned with the latest WHO guidelines. The EU Zero Pollution Action Plan (European Commission 2021) demands for a reduction of health impacts caused by air pollutants by 55 % for 2030 compared to 2005. Actually, this is not far away from the objective of the NEC Directive aiming at a reduction of 50 % in the same period. As shown in chapter 2.2, this ambition level will by far not reach the WHO AQG Levels. To align future air quality with the WHO AQG as far as feasible by cost-efficient means, we propose to investigate further measures beyond the ERC. Moreover, while the following recommendations are based on available abatement measures, we consider it necessary to continuously improve air quality beyond 2030, aiming to achieve the WHO AQG levels by a step-by-step reduction of the proposed Air Quality Standards (AQS).

Besides agreeing a level of ambition, it must be considered, whether an AQS should be set as a binding limit value or a non-binding target value. Where no sufficient evidence for adverse health effects is available for a pollutant, mandatory monitoring could be a first regulatory step.

Moreover, the obligation to attain a binding limit value must go along with available cost-efficient measures on Member State (MS) level. For pollutants where only global action can result in significant improvements, a target value seems more appropriate.

As has been shown an improvement of air quality can be expected in Germany due to the implementation of the National Air Pollution Control Programmes aiming to fulfil the ERC according to the NEC Directive.

Progress and extent of expected transformations of the energy and the transport sector will significantly influence the development of air quality. In the following we evaluate for each respective pollutant which Interim Target will be attainable in a specific target year. As a national authority we restrict our analysis to the urban and regional background. Local hot spots may show substantially higher concentrations. Our analysis has indicated that apart from PM_{10}, CO and SO_2 the latest WHO AQG Levels cannot be met everywhere in Germany at background locations by cost-efficient measures in the next decade.

In this paper we focus our recommendations just on long-term AQS, noting that also short term-AQS are justified from a health standpoint. Moreover, short-term AQS can support public information and awareness raising. Beside this, they may trigger mitigation measures on a local scale at locations with recurrent short-term concentration peaks, e. g. situations with short-term elevated concentrations in residential areas caused by domestic heating. These concentration peaks often do not cause an exceedance of air quality standards based on annual means, but still have negative health impacts on residents. In such situations short-term AQS have the additional advantage that they do not require monitoring during a whole year but exceedances can be assessed by temporary measurements. By using a regional CTM with a coarse spatial resolution we cannot give recommendations which Interim Targets or AQG Levels for short-term standards might be attainable.

2.1 Binding long-term air quality standards

Nitrogen dioxide – NO_2

Elevated NO_2-concentrations occur mainly due to vehicle emissions in urban areas. Though tightening of emission standards is within the responsibility of the EU, local authorities may implement measures which substantially contribute to the reduction of NO_2-levels, e. g. low emissions zones. Therefore, and with a view to the expected improvement by implementing the NEC Directive, we consider it appropriate to lay down a binding long-term limit value in the range of Interim Target 3 for NO_2-concentrations (20 µg/m³) in the urban background to reduce the still relevant risk to human health.

Particulate Matter – PM

Compared to NO_2 a notably higher burden of disease can be attributed to particulate matter, i. e. $PM_{2.5}$ as well as PM_{10}. Therefore, in spite of a considerable transboundary transport in particular of $PM_{2.5}$, we consider a binding long-term limit value in the range of Interim Target 4 for $PM_{2.5}$ (10 µg/m³) as feasible. For PM_{10} the AQG Level (15 µg/m³) would be attainable with additional effort, e. g. addressing residential heating by wood firing. In particular in larger MS such as Germany limit values in addition to national ERC help to avoid areas with elevated concentrations.

The classic concept of limit values, not to be exceeded at any locations where members of the public do have access, mainly aims to reduce concentrations at hot spots. Some local air quality plans tackle concentration peaks with single measures acting on confined street sections, e. g. the installation of high-volume air filters. From these measures the health of only a limited number of persons will benefit. To avoid this shortcoming in a future AAQD, we propose – in addition to a limit value applying in the urban and regional background – binding obligations to reduce the overall exposure of humans to air pollution (see also Hoffmann et al. 2021). To better allocate responsibilities and address the most relevant sources the already existing average exposure indicator (AEI) should be modified to aim at the reduction of the average exposure to PM of the whole population on a more regional level instead of only on the national level.

Carbon monoxide and sulphur dioxide – CO and SO_2

We propose to set the WHO AQG Levels (CO 4 mg/m³; SO_2 40 µg/m³) as future limit values. They are expected to be met everywhere in Germany by 2030.

Benzo[α]pyrene – BaP

Following the WHO AQG we propose to establish a binding limit value for BaP instead of setting only a non-binding target value. Considering the abatement potential in residential heating a concentration well below the current target value of 1 ng/m³ seems attainable.

2.2 Non-binding long-term target values

Ozone

Current ozone levels in Germany only meet the WHO Interim Target 1 at most monitoring sites. Ozone mitigation would therefore substantially contribute to the reduction of health risks from air pollution. However, ozone being a secondary pollutant its abatement depends on the reduction of its precursors. As shown in Chapter 2.2 NO_2- and NMVOC-mitigation as expected from the NEC Directive hardly improve the situation until 2030. From modelling studies it becomes evident, that biogenic VOC and methane are the most relevant precursors in Europe. E. g. Butler et al. (2020) showed in a recent study that about 30 per cent of surface ozone in Germany originates from methane, but most of this ozone fraction (84 per cent) is produced outside of Europe. Yet, for both these precursors it is very difficult to develop further national abatement strategies to substantially reduce the ozone levels.

With a view to this we recommend to set an ambitious, but non-binding target value for ozone preferably stricter than Interim Target 1 (100 µg/m³). The need for a global abatement strategy for methane should be reflected in the objectives of a revised AAQD.

2.3 Implementation of AQS

We propose that – as in the current AAQD – all air quality standards should be related to a fixed year. To align air quality planning with the ERC under the NEC Directive we recommend the year 2030 as a basis, but also a perspective should be given for the following decade.

To evaluate the foreseen measures with respective to attaining the AQS, we recommend that regular air quality plans should be drafted. We consider the use of a CTM, both on the regional and locale scale with the regional CTM using emission data from the national inventories, as mandatory in this context.

Air quality plans should be set up right from the implementation date of a new AAQD, ensuring that all necessary measures are taken to meet the limit values when they come into force.

2.4 Gaining scientific evidence for future AQS

Ultrafine Particles and Black Carbon are two examples of air pollutants, where significant knowledge gaps still exist and therefore setting up target or limit values would not be scientifically sound. Hence, a Zero Pollution Ambition Cycle (ZPAC) developed by Conrad et al. (2021) in response to the EU Zero Pollution Action Plan (European Commission 2021) cannot be fully implemented. The integrated ZPAC contains the identification of main pressures on health and the environment through monitoring, selection and implementation of appropriate measures and the control of success including the assessment of policy effectiveness through monitoring and the examination of trends of pollution.

Ultrafine Particles – UFP

Impact studies indicate that UFP in outdoor air pose a risk to human health. However, this risk cannot be equated with exposure to fine dusts in general (PM_{10} or $PM_{2.5}$). UFP penetrate deeper than the larger particles into the lungs, the brain (via the olfactory nerve) and presumably even directly into the blood circulation system via the inhalation uptake pathway. This results in a possible distribution and deposition of UFP in various organs. In summary, there is a need for epidemiological studies on the different short- and especially long-term effects of UFP, where the high spatio-temporal variability of UFP is considered and a high comparability of measurements and studies is given (Morawska et al., 2019; Ohlwein et al., 2019; Rückerl et al., 2011; Schraufnagel, 2020).

In its latest AQG also WHO considers UFP to be a parameter of particular interest, even if the current state of evidence does not yet permit a recommendation for a Guideline Level. WHO advises the further development and permanent use of standardised measurement methods that enable a meaningful comparison to be made between the results of different studies. This would also enable a better characterisation of exposure to UFP and a verification of the effects of UFP in epidemiological studies.

UFP are currently mostly measured and described by the total number of particles and the number/size distribution. Optical and gravimetric methods, as usually applied for the determination of fine dust, are not applicable. In order to improve the comparability and standardisation of data, the range of the UFP size detected by emission and concentration measurements should be harmonised. In particular a traceability to existent standards is needed (CEN 2020a, 2017, 2016).

In order to obtain an improved assessment of the exposure to UFP in the higher densely populated regions and its health consequences, we recommend an obligation to establish monitoring sites in the urban background and in metropolitan regions (>1 million inhabitants). Moreover, source-related measurements to quantify specific sources should be enhanced to be able to introduce effective source-specific mitigation strategies. This is because the main emitters have been identified in principle but only partially quantified, and others such as shipping and small combustion installations have so far only been insufficiently characterised and assessed. To improve the attribution of polluters, further development of dispersion models should be required, e. g. with regard to the description of particle formation and the incorporation of non-volatile and volatile components, as well as the compilation of emission inventories of UFP and their precursors.

Black Carbon – BC

Impact studies show that BC in outdoor air pose a risk to human health. In its latest AQG, also WHO considers Black Carbon to be a parameter of particular relevance without proposing a Guideline Level (WHO 2021).

Currently, from a health point of view, effects caused by BC cannot be clearly distinguished from those by PM_{10} or $PM_{2.5}$. However, epidemiological studies show a higher robustness of the models and a stronger effect (especially for short-term effects) when using BC as an assessment and prediction parameter, instead of $PM_{2.5}$. Multi-parameter models also show a clear influence when adjusting the $PM_{2.5}$ model with BC, but a significantly smaller influence when adjusting the BC model with $PM_{2.5}$. Thus, BC represents an independent parameter providing additional information on health effects, especially of primary combustion particles that are not necessarily addressed by regulating PM (Janssen et al. 2012).

From a metrological point of view, two methods, a thermal-optical and an optical (transmission, absorption) method, are widely used. The thermal-optical method analyse the EC/OC mass content collected on filters and has to be applied in regulatory monitoring (CEN 2017). The advantage of the optical method is an almost real-time recording of a BC signal converted to eBC (equivalent BC) mass values. In the least years many studies demonstrated that both methods show reliable data and are comparable to each other.

In order to obtain an improved assessment of the exposure to BC we consider it necessary to establish mandatory BC monitoring sites primarily in densely populated regions analogue to the proposed monitoring of UFP.

3 Ambitious limit values require changes in governance

As shown above a better alignment of the EU AQS with the latest WHO AQG should be a key element of a revised AAQD. A modified governance structure would be of high importance to reach this aim.

Under the current AAQD, MS and their regions are responsible for compliance with limit values. However, air pollution can often only be reduced slightly by local measures alone:

- Most of the emission source legislation is now regulated at EU level (IED, MCPD, eco design regulations, emissions legislation for cars, light and heavy commercial vehicles, mobile machines and devices) limiting the legislative competences of a MS and its regions to adopt provisions at national/local level to improve air quality.
- A large share of emission sources causing high local concentrations is often outside jurisdiction of responsible entity (particularly where the relative importance of the background pollution caused over a wide area increases due to measures taken). But also for emission sources within their jurisdiction local authorities have very few instruments to improve air quality. Often the only remaining option would be banning or reducing activities of the local population.

The future AAQD should therefore include provisions that ensure both coherence and transparency between different levels of governance, e. g. a provision requiring Union institutions and Member States to cooperate and take the necessary measures at Union and national level. Similar challenges in Climate Policy have been resolved in the EU Climate Law with a provision resulting in a joint legal responsibility of EU institutions and Member States, within their respective competences. In general, the Zero Pollution Ambition provides a chance for bringing forward cross-regulatory approaches. Therefore, UBA has developed the so called Zero Pollution Ambition Cycle for a cross-regulatory approach (Conrad et al. (2021)).

On the practical side a reporting mechanism, which might be implemented as an additional data flow within the existing Air Quality e-Reporting, ensuring early transparency between all governance levels is needed to enable e. g. EU institutions to foresee which changes in EU emission source legislation are necessary.

4 Protecting ecosystems from ammonia and ozone

Besides reducing the risk to human health, the revised AAQD should keep the objective to protect ecosystems from air pollutants. The current legislation sets air quality standards for SO_x and NO_x which are still in line with the state of science. However, ammonia, despite its known impact on ecosystems, is not yet regulated in the AAQD. For ozone latest scientific evidence suggests a change of metrics.

4.1 Ammonia – making monitoring mandatory

Motivation

Ammonia impacts ecosystems and contributes to the loss of biodiversity (Bobbink & Hettelingh 2011, Krupa 2003). In addition, it is a key precursor in the formation of $PM_{2.5}$ (CLRTAP 2021). It is likely that critical levels of ammonia as set by Convention on Long-Range Transboundary Air Pollution (CLTRAP) (Cape et al. 2009, CLRTAP 2017) are exceeded for a substantial number of protected ecosystems in Europe (Schaap et al. 2018). Those impacts cause costs in the European health system and for ecosystem restoration (CLRTAP 2021, Gu et al. 2021, Stokstad 2014).

In contrast to other major air pollutants, measurements at EMEP background sites show that on average ammonia concentrations in Europe have been rather increasing in the last two decades (Fagerli et al. 2021). Following the current EU ERC (European Union 2016) and the preparatory emission projections presented by IIASA at the first stakeholder meeting to inform the process on Air Quality: Revision of EU Rules (Klimont et al. 2021), for the next three decades, ammonia emissions will be reduced much less than e. g. those of nitrogen oxides, making ammonia and the agricultural sector an even more prominent source of reactive nitrogen and $PM_{2.5}$ in the future.

Effective ammonia emission reduction strategies require the knowledge of the impact those reductions have on both ecosystems and $PM_{2.5}$-formation. Directive (EU) 2016/2284 on the reduction of national emissions of certain atmospheric pollutants requires national emission reductions of ammonia. Complementary, a better local and regional understanding of the spatial and temporal distribution of atmospheric ammonia are needed to lay a profound basis for regional ammonia emission reduction strategies and help to improve models needed to support appropriate strategy development.

Objective

According to the EMEP monitoring programme long-term ammonia measurements take place at a limited number of background sites (Fagerli et al. 2021). However, despite obvious threats and increasing trends underpinning the need for effective reduction strategies, no impact-oriented approach exists for assessing ammonia concentrations across Europe. Only a few MS have implemented specific ammonia monitoring networks for ecosystems (e. g. Nordijk et al. 2020). With a view to the future, the monitoring of ammonia should be made a mandatory element of a revised Directive 2008/50/EC. In that way EU legislation will

- ► allow for regional and local assessments to support reduction strategies on that level,
- ► supplement and foster already existing knowledge on effect levels for ammonia for ecosystem protection provided by CLTRAP,
- ► improve the understanding of ammonia and the formation of $PM_{2.5}$ in the atmosphere for better implementation in atmospheric models, which form an important basis of EU Clean Air policies.

By incorporating ammonia into the revised AAQD, the basis will be laid for a common EU-wide standardisation and agreement on ammonia related effect levels for ecosystem protection, reference methods for the measurements and siting criteria.

Proposal for a monitoring concept

In the revised Directive 2008/50/EC ammonia monitoring should be mandatory. Data and relevant metadata should be reported together with other mandatory pollutant data to the European Commission each year. They should be evaluated against the critical levels set by Convention on Long-range Transboundary Air Pollution (CLRTAP).

Most preferably, chosen sites would represent nitrogen sensitive ecosystems, covering the most relevant ecosystems within regions of similar ammonia pollution. Where appropriate, sites would be established in Natura2000 areas and/or make use of existing monitoring stations if possible such as EMEP or AAQD stations, while both affected and non-affected ecosystems should be chosen. Affected ecosystems may be identified by their vicinity to emission sources or through evidence of high ammonia levels in atmospheric models. For evaluation of ammonia effect levels, in addition vegetation surveys at both affected and non-affected sites would be recommended. The number and location of selected sites are to support the above outlined objectives with measurements employed in all prevailing ecosystems types, covering regions of highest ammonia emissions and reflecting the geographic and climatic variability in each MS.

The use of a standard measurement methodology is key for the comparability between sites across Europe. Measurements should cover the entire annual cycle, using passive samplers (CEN 2020b) or denuders (VDI 2010) with a sampling rate of preferably two weeks or less and not exceeding four weeks. Intercalibrations with other methods and between MS should be part of the quality control. Where possible, supporting measurements of particulate ammonium can be established to monitor the overall effect of reduced nitrogen on ecosystems.

To achieve the above-mentioned objectives the monitoring data should be continuously reviewed, e. g. by a group of experts from MS, which might also propose improvements of the monitoring concept.

4.2 Ozone – introducing a new metric

Motivation and objective

Besides adverse effects on human health (see Chapter 3.2), there is clear evidence that ongoing exposure will lead to chronic ozone effects on vegetation and that these effects already occur at comparably low concentrations. In Directive 2008/50/EU the AOT40 is used as metric for an AQS, which reflects only the exposure of vegetation to elevated ozone concentrations above 40 ppb accumulated in one hour. However, the relevant effects in plants are caused by the accumulated flux of ozone via their stomata. Therefore, International Cooperative Programme (ICP) Vegetation, a CLRTAP body of scientific experts from several countries, agrees that the AOT40 approach can lead to false evaluation of the real risks in many cases. The application of the flux-based method (see Box) is recommended in the Modelling and Mapping Manual of CLRTAP, Chapter 3 (CLRTAP 2017). The NEC Directive in Article 9 and 10/ Annex V already takes this account in recommending monitoring and reporting of the exceedance of flux based Critical Levels as a key indicator for risk assessment of tropospheric ozone in addition to monitoring of visible injury of needles and leaves (see Annex V, c). But the NEC Directive does not contain any target values to be met or clear requirements for the monitoring network.

Concept of an ozone flux-based air quality standard

Harmful effects of ozone to vegetation are not only determined by ozone concentrations above a certain threshold value, but are also strongly influenced by meteorological conditions and soil moisture. These factors are included in the concept of the "Phytotoxic Ozone Dose" (POD), a flux-based approach developed in ICP Vegetation. By doing so, the POD is much more relevant to ecotoxicological effects than the AOT concept, according to present knowledge (Hayes et al. 2007, Braun et al 2014 corroborating increment reduction rates of trees in Harmens and Mills 2012; Sicard et al. 2016, Bender et al. 2015).

The risk assessment based on Critical Levels related to POD as derived by ICP Vegetation compares effects at present, e. g. yield losses, with the pre-industrial pollution situation (ambient ozone concentration assumed to be 10 ppb). Thus, to reach non-exceedance of the Critical Level could at the best be understood as a long-term objective.

Aiming at an attainable AQS, it would be necessary to set interim targets with a lower ambition. This would mean that e. g. yield losses still can occur but be smaller than today or that quality indicators are still affected, but to a lesser extent. E. g. a method has been derived (Grünhage et al. 2018, VDI 2310 part 6), relating effects to the situation up to the 1980s (concentration levels approximately factor 0.6 to the current situation in Germany, i. e. 15 - 20 ppb). The approach has been laid down in VDI 2010 Part 6, and could be considered suitable for Central Europe. It would be recommendable, however, to look for data from more sites over Europe for a broader evaluation of the method.

Table 3 Target Values for PODY based on Table 2 in VDI 2310 Part 6 (slightly changed)

Receptor	Effect parameter	Value of Y (in PODY)	Critical Level CLe PODYSpec[2] [mmol m-² PLA]	Target Value TV PODYSpec[3] [mmol m-² PLA]
Winter wheat	Grain yield	6	1,3	3
Beech	Annual growth of whole-tree biomass	1	5,2	13
Spruce	Annual growth of whole-tree biomass	1	9,2	15
Species-rich grassland	Number of flowers (indication of biodiversity, ecosystem services)	1	6,6	11

Alternatively, an interim target could be defined by setting percentages of the number of sites with no Critical Level exceedance. This would require clear regulations, how sites are to be selected. Another alternative could be to set an aspired percentage of reduction of the extend of exceedance [mmol/m² PLA].

It should also be considered whether the POD concept could be simplified by reducing the number of effects endpoints (several plant species/vegetation types, each having critical levels for yield, quality or biodiversity, resp., see Tab III.10, Tab III. 12, Tab III.14 in Manual Chapter 3). E.g. a most sensitive vegetation could be used as general indicator in order to protect all ecosystems.

[2] CLe PODYSpec = Critical Level based on the species specific Phytotoxic Ozone Dose with y indicating the detoxification potential of the species

[3] TV PODYSpec = Target Value based on the species specific POD of a site in the estimated ozone pollution situation of 1980s with y indicating the detoxification potential of the species

Proposal for regulating ozone

The revised Air Quality Directive should continue considering the protection of ecosystems by setting a target value for ozone effects on vegetation. However, according to the latest scientific evidence we recommend that vegetation damage by ozone should be assessed by using the POD instead of an AOT-concept. Different approaches could be considered in doing so (see Box).

Using the POD-approach would also require to revisit the monitoring concept. Although in Annex V of the NEC Directive (EU) 2016/2284 one of the recommended indicators is the exceedance of flux-based Critical Levels of ozone, no clear requirements for the monitoring network and parameters to be assessed are provided. This means, however, that so far neither the NEC Directive nor the current AAQD set any AQS related to the assessment of POD. In order to enable evaluation of attainment of POD-based target values, which is comparable between Member States, clear criteria for representativeness, completeness and quality for the monitoring network should be fixed in the AAQD.

The air quality measurement networks for tropospheric ozone should be evaluated with respect to their representativity for effects on ecosystems, completeness and quality of measurement data for calculations of the POD. Complete yearly datasets of hourly ozone concentrations and meteorological data (temperature, humidity, radiation, partial pressure, wind speed) would be needed. We recommend to lay down clear rules and appropriate geostatistical methods for ensuring sufficient representativeness, completeness and quality of measurement networks' datasets and for options to replace measurement data by modelled data in the reporting by the MS with respect to ozone effects on vegetation.

The experience gained from several years of measurement across the EU and reporting obligations should be used to derive POD-related air quality standards in a second step (see Box Concept of an ozone flux-based air quality standard).

5 Improving the assessment of compliance with long- and short-term air quality standards

Stemming basically from the year 1996 the current AAQD allows for monitoring as the only appropriate method to assess compliance above the upper assessment threshold. In the past decades though, modelling has substantially improved and should be considered as an additional tool, thereby enabling air quality assessment for the whole area of a MS.

5.1 Current regulation

According to the Air Quality Directive 2008/50/EC, 'sampling points directed at the protection of human health shall be sited in such a way as to provide data' on areas 'where the highest concentrations occur to which the population is likely to be directly or indirectly exposed for a period which is significant in relation to the averaging period of the limit value(s)' (Annex III B 1). With a few exceptions given in Annex III A, this requirement establishes a hotspot concept where limit values for the protection of human health have to be attained not only at background sites but also at traffic and industrial sites with high concentrations of air pollutants. If sampling points are located at the real hotspots, compliance with limit values at these hotspots ensures compliance everywhere.

Furthermore, modelling techniques may only be used to assess ambient air quality below the upper assessment threshold. Otherwise, measurements are mandatory for air quality assessment.

5.2 Motivation for a new assessment concept

Air pollutants show typical spatial-temporal patterns with peak concentrations occurring on short time scales (minutes to hours) and often being confined to specific locations (m^2 to km^2). These peak concentrations at hotspots are important to asses short-term exposure of humans to air pollutants, but they are not necessarily representative for long-term exposure. Therefore, the assessment of short-term air quality standards (hourly or daily means) should be designed to address short-term exposure, whereas the assessment of long-term air quality standards (annual means) and exposure concentration obligations should reflect long-term exposure.

Table 4: Assessment of different spatio-temporal obligations

short-term air quality standard	representing short-term exposure	assessment by concentration values representative for short time periods (hours to one day) on a pollutant-dependent spatial scale* at fixed measurement sites
long-term air quality standard	representing long-term exposure	assessment by average values representative for annual concentrations at fixed measurement sites
exposure concentration obligation	representing long-term exposure	assessment by average values representative for annual concentrations on a km^2-scale

* for nitrogen dioxide an appropriate spatial scale may be a few square metres, whereas for ozone an appropriate spatial scale may be tens of square kilometres.

5.3 Assessment of short- and long-term air quality standards

Improvements in modelling techniques offer the opportunity to broaden their possible applications for air quality assessment. Whether short-term air quality standards are met should still be assessed by measurements at hotspots, since short-term fluctuations of concentrations and short-term concentration peaks caused by peaks of emissions from nearby sources cannot be reliably resolved by microscale models. However, regional CTM may be used to support the assessment of long-term air quality standards by measurements or may even be used to provide an area-wide assessment of exposure concentration obligations, provided appropriate quality management is applied and emission data with a necessary spatial and temporal resolution are available.

Widening the possibilities to use CTM for air quality assessment should not lead to a reduction of the number of existing measurement sites. Measured data are important to assess both air quality and the quality of outputs from CTM and they can also be used to apply data assimilation techniques to improve model results. Nevertheless, the siting of measurement sites may be redesigned to meet requirements arising from model evaluation or data assimilation.

The assessment of air quality with respect to exposure concentration obligations should be representative for long-term exposure of humans. To perform this kind of assessment, different approaches using measurements or CTM or combination of both are possible:

- **Assessment by measurements**
  An assessment based on measurements can be conducted by averaging data from suitable (urban) background sites.
- **Assessment with CTM**
  - Assessment is based on the consideration of each grid cell (i. e. the modelled annual concentration in each grid cell has to meet the obligation)
  - Assessment is based on averages over different grid cells, e. g. average over all grid cells of a certain urban agglomeration
  - Averages over different grid cells may also be calculated as population-weighted averages
- **Combination of CTM and measurements**
  An assessment based on CTM may serve as a preliminary assessment. If the modelled concentrations show exceedances of exposure concentration obligations, the exceedance has to be confirmed by measurements during the next year.

6 Supersites: Aiming at a better understanding of air pollution

As before the foremost objective of the upcoming AAQD will be to improve air quality by respecting AQS for specific pollutants which are known to pose a risk to human health and ecosystems and which are in the centre of monitoring activities. However, a limited number of so-called Supersites would help generate knowledge for an improved scientific understanding of air pollution and better targeted air quality policies in the future.

6.1 Extended monitoring at Supersites as a basis for better air quality policy

Beside ensuring compliance, knowledge on the generation, transport and aging of regulated and un-regulated air pollutants, as well as on precursors, deposition processes and sources is needed to understand air pollution and address health and ecosystem effects properly. Therefore, systematically collecting this information should be reflected in legislation. Such information would also enhance the development, improvement and mandatory validation of CTM. A combination of modelling and in situ monitoring by so-called Supersites would bridge high quality single data point measurements and modelling specifically to gain information for area-wide concentrations and thereby exposure (see chapter 3.1).

The aim is to provide additional high quality and objective-specific data with higher time resolution that can be used for thorough evaluation and improvement of air quality monitoring, characterization and models.

Supersites should improve the general air quality characterisations and monitoring of a wide set of parameters, thereby allowing for the calculation of e. g. mass balances, trend analysis and source identification. For the application, evaluation and improvement of CTM Supersites could provide measurement data with high time resolution that allow diagnostic evaluation of the model robustness and the implementation of transport and chemical processes. An evaluation of emissions and model parameterisations regarding particle nucleation and growth processes would become feasible, leading to an improved understanding of atmospheric process. Current – sometimes relatively high – uncertainties could be improved by testing with observed data.

Overall the strength of Supersites would be the combined measurement of "common parameters" (e. g. PM, ozone), precursor observations like volatile organic compounds and chemically active parameters (e. g. ammonia).

6.2 Characteristics of a Supersite programme

Supersites should be designed as a platform for additional air quality related projects, questions and measurements that are addressing general improvement of data quality and models as well investigating specific scientific questions. They should cover the mandatory monitoring related to AQS as well. This could be done by a mixture of permanent and temporary campaign measurements.

Each Supersite should be placed, equipped and installed to address process and/or source-receptor issues in the context of exposure, health risks or ecosystem protection. Therefore, supersites are recommended to be placed in geographic areas with a range of representative characteristic of air quality component source-receptor, exposure, environmental and health risk situations. Supersite monitoring can be newly implemented as well as the profile of existing (monitoring network) stations can be updated. The latter would be in particular preferable if trend analyses is also one of the objectives. Newly implemented sites are placed ideally close to existing assessment programmes, e. g. cohorts, existing networks etc.

Due to the cost and high management expenditure it would not be feasible to have a nationwide Supersite monitoring network addressing all objectives. We therefore recommend Supersites in number, profile and location specifically related to member states' characteristic and challenges in air quality improvement. These challenges could be caused by e. g. industry, agriculture, traffic, etc.

The air quality monitoring by Supersites should, besides the already existing monitoring tasks, aim at special parameters useful for model validation, source apportionment and process understanding. Mandatory parameters should be: Meteorology (air temperature, precipitation, relative humidity, radiation), wet and dry deposition (particles and their composition) and gas analysis inclusive chemistry (metals, ions, in-organics, PM, VOCs, POPs). For the group components of VOC, POP and inorganic elements common minimum requirements should be agreed while the extended individual monitoring programme has to be adapted to the objectives and designation of the Supersite. The same is true for the time resolution of the monitored data.

For all parameters a quality assurance has to be implemented, respectively to be adjusted. As long as they are in line with the directives, existing quality assurance of networks like ICOS or ACTRIS are open to use to make sure station data sets are comparable to existing networks and monitoring data.

All data from Supersites should be reported annually, including descriptive statistics, trend analysis, source apportionment and data interpretation. To achieve the objective of supporting air quality policies, the AQUILA network might be encouraged to draw conclusions from the data gained at Supersites.

List of abbreviations

AAQD	Ambient Air Quality Directive
AEI	Average Exposure Indicator
AOT	Concentration Accumulated Over a Threshold, e. g. AOT 40 (threshold = 40 ppb)
AQG	Air Quality Guidelines
AQS	Air quality standard
BC	Black Carbon
CLRTAP	Convention on Long-range Transboundary Air Pollution
CTM	chemical transport model
EC	Elemental Carbon
EMEP	Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe
ERC	emission reduction commitments
GRETA	Gridding Emission Tool for ArcGIS
ICP	International Cooperative Programme under the CLRTAP
MS	Member State
PLA	Projected Leaf Area
PM	Particulate Matter
POD	Phytotoxic Ozone Dose above a flux threshold of Y nmol m^2 PLA s^{-1} accumulated over a stated time period during daylight hours
UBA	Umweltbundesamt - German Environment Agency
UFP	Ultrafine Particles
WHO	World Health Organisation
ZPAC	Zero Pollution Ambition Cycle

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