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Review of internationally proposed critical levels for ammonia

Proceedings of an Expert Workshop held in Dessau and online on 28/29 March 2022

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Abbreviations

AAQD	Ambient Air Quality Directive
BImSchV	Bundesimmissionsschutzverordnung
CCE	Coordination Centre for Effects
CEH	Centre for Ecology and Hydrology (UK, several units)
CEN	Comité Européen de Normalisation
CLe	Critical Level for Air Pollutant Concentration, µg m^{-3}
CLo	Critical Load for Deposition, kg ha^{-1} yr^{-1}
CLRTAP	UNECE Convention on Long-Range Transboundary Air Pollution
CRDS	Cavity Ring-Down analyzers
DEFRA	Department for Environment, Food & Rural Affairs
DOAS	Differential Optical Absorption Spectroscopy
EMEP	The co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe, European Monitoring and Evaluation Programme
IED	Industrial Emissions Directive
INI	International Nitrogen Initiative
INRA	Institut National de la Recherche Agronomique (France, several units)
ICP	International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects e.g. on Forests
IPCC	Intergovernmental Panel on Climate Change
IPPC	Integrated Pollution Prevention and Control
LANUV	Landesamt für Natur, Umwelt und Verbraucherschutz (LANUV) Nordrhein-Westfalen
LfU	Bayerisches Landesamt für Umwelt
LRTAP	UNECE Convention on Long-range Transboundary Air Pollution
LTER	Long-Term Ecological Research Network
LUBW	Landesanstalt für Umwelt Baden-Württemberg
MAK	Maximale Arbeitsplatz Konzentration (permissible workplace concentration)
NEC	National Emission Reduction Commitments Directive 2016/2284/EU
NERC	Natural Environment Research Council
OGS	Optical gas standard
PM	Particulate Matter PM_{10}, $PM_{2.5}$
PRTR	Pollutant Release and Transfer Register of the EU
QC-LAS	Quantum Cascade Laser Absorption Spectroscopy
ReGaS	Reference gas standard
RIVM	Rijksinstituut voor Volksgezondheid en Milieu (NL, Bilthoven)
TA Luft	Technische Anleitung zur Reinhaltung der Luft
UBA	Umweltbundesamt (German Environment Agency)
UNECE	United Nations Economic Commission for Europe
VDI	Verein Deutscher Ingenieure (The Association of German Engineers)
VMM	Vlaamse Milieumaatschappij (Flanders Environment Agency)

1 Background

Air pollution from ammonia (NH_3) is relevant for human health and ecosystems. The gas is easily converted into NH_4^+, which can act as a plant nutrient, either by assimilation via the plant shoot (after entering the stomata) or after deposition to the ground and the subsequent uptake by the roots (Sutton et al. 1993; Fangmeier et al. 1994). Together with the oxidized forms of nitrogen, it is significantly contributing to the widespread eutrophication and acidification of ecosystems. As a result, the biodiversity of ecosystems is threatened at the expense of slow-growing nitrogen-sensitive, often rare and protected species (Bobbink et al. 2010).

Approximately 95 % of the ammonia (NH_3) emissions stem from agriculture, especially from livestock farming and the application of slurry or mineral fertilizers (Umweltbundesamt 2021). The odour threshold of ammonia is 5 ppm (3.8 mg m^{-3}) and the permissible workplace concentration (MAK) is 20 ppm (15 mg m^{-3}). Inside stables and after slurry application in the field, peak concentrations can be above 1 ppm (760 µg m^{-3}), whereas own measurements have shown that outdoors and close to stables, mean levels will still be around 50 µg m^{-3}. Slightly elevated concentrations of the gas in the range of 10 µg m^{-3} can also be detected near waste-water treatment plants. Major roads can act as line sources, but concentrations drop away from the edge of the road (Cape et al. 2004; Bell et al. 2016). In remote areas, ammonia concentrations are often below 1 µg m^{-3} (Umweltbundesamt 2018), while the presence and excrements of animals (urea and uretic acid) and the degradation of plants will elevate levels locally.

Due to its alkaline character, ammonia is rapidly reacting with acids (e.g. sulphuric and nitric acid) creating ammonium salts like $(NH_4)_2SO_4$ and NH_4NO_3, which possess long atmospheric lifetimes as secondary aerosols (PM 2.5) (Fowler et al. 2009). However, the strong reduction of the acidic air pollutants in the last decades has led to more alkaline atmospheric conditions and an increase in the lifetime of ammonia due to the reduced availability of H_2SO_4 to react with NH_3 (Sutton et al. 2020).

To protect European ecosystems from eutrophication, ammonia emissions will have to be reduced significantly in the EU according to the national emission ceilings laid down by the updated NEC Directive 2016/2284/EU and the 2020/2030 national emission reduction commitments. Most countries are currently developing and applying methods to assess nitrogen effects on ecosystems and vegetation with critical loads, in contrast critical levels are only used sporadically in the UK, Denmark and Germany within the framework of licensing new installations (Reinds et al. 2019).

Critical levels (CLe) describe the concentration of a pollutant above which direct adverse effects on receptors such as individual plants or natural ecosystems may occur. CLe for ammonia had been last discussed in the framework of the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) at a Workshop on Atmospheric Ammonia (UNECE 2007). New experimental evidence was provided including from experiments at Whim Bog in Scotland (Cape et al. 2009) suggesting that the long-term critical level of ammonia needed to be reduced to 3 µg m^{-3} for higher and 1 µg m^{-3} for lower plants (i.e. lichens and mosses), respectively. The CLRTAP accepted these recommendations replacing existing the previous CLe of 8 µg m^{-3} (Ashmore and Wilson 1994; UNECE 2007).

More than ten years after the recommendation of updated CLRTAP critical levels for ammonia in the Mapping Manual, new findings on the effects of ammonia on vegetation have been discussed on a workshop prepared by the CCE and the German Environment Agency (UBA) as a lead. The workshop was held at Dessau on March 28/29 March 2022 and offered the opportunity to exchange information on national or regional programs which have been set up for the monitoring of ammonia in sensitive habitats. Scientists dealing with research on effects of ammonia on vegetation and ecosystems and those involved in the monitoring of ammonia in the environment were also asked to present their recent research.

The workshop was part of the current work plan for the year 2022-2023 of CLRTAP (1.1.1.22) and was organized in the framework of a R&D Project financed by the German Environment Agency (UBA). Due to the corona restrictions, only 15 participants were able to attend in person but more than 100 people were following the live stream via a WebEx meeting platform. Participants represented several European countries and the USA. In total, 19 talks were given from presenters from nine countries.

In the present workshop proceedings, we include a review of the latest findings and give summaries of the presentations given by the workshop participants.

2 An overview of ammonia research

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In order to evaluate the validity of current critical levels and to prepare our own fumigation study (see Kösler & Franzaring, chapter 4.2.2), we screened the literature on ammonia effects from the last twelve years, focussing primarily on original publications which cited Cape et al. (2009). Nevertheless, we also reviewed important publications prior to 2009 in present background paper including fumigation studies from the 1970-1990s, that are helpful to better differentiate between phytotoxic vs. chronic effects and to address key points that need to be respected in fumigation experiments. Besides the peer-reviewed literature, we also included various reports from studies that monitored ammonia in regional and national projects.

2.1 Research in the early years

Experimental exposure of plants to ammonia in the very early years was mainly based on the use of pure (technical) NH_3 supplied from gas flasks to the air inlet of small closed fumigation chambers or continuously-stirred tank reactors (CSTR). Very high concentrations were applied initially to examine whether the growth of crop plants, e.g. sunflowers, could be stimulated in growth by nitrogen containing gases (Faller 1972; Ewert 1979; Temple et al. 1979; Farquhar et al. 1980). In these short-term experiments with young plants, phytotoxic responses (foliar injury and growth reductions) were observed only at concentrations above 800 µg m^{-3}, depending on the plant species and the soil nitrogen supply. It had also been suggested early that fertilized crops and older plants do not take up ammonia any more at concentrations below 4 µg m^{3}, but release the gas during senescence (Cowling and Lockyer 1981).

Unfortunately, many of the early and more recent studies did not mention sufficient details how the ammonia fumigation was controlled, whether constant levels were achieved and how the realized concentrations were determined. Optimally, the relationships between the inlet and the outlet NH_3 concentrations would have to be monitored constantly (Jones et al. 2007). On the other hand, it will also be necessary to give a better account of how the test plants (receptors) were grown and what nutrient supply they received prior and during the fumigation. An overview of published ammonia fumigation experiments prior to 2009 is given in Table 1.

Dry deposition, bi-directional fluxes and compensation points of ammonia have been addressed early by Scottish, Dutch and Danish scientists (e.g. Sutton et al. 1993; Duyzer 1994 and Husted and Schjørring 1996) and have been parameterised in air-vegetation models later (Loubet et al. 2001; Nemitz et al. 2004). The fluxes are driven by changes to the equilibrium between the ammonia concentrations in the air and in the sub-stomatal cavity of plant leaves and depend on the air/leaf temperature, stomatal and cuticular resistances, seasonal and diurnal changes in stomatal opening, plant water status, ammonium (NH_4^+) concentration (nutrient status) and pH of the apoplastic solution.

Farquhar et al. (1980) and van Hove et al. (1987) obtained compensation points of around 5 µg m^{-3} in crop plants, but suggest that in oligotrophic vegetation, e.g. peatlands, heath and dunes, the uptake of ammonia may occur at lower concentrations (e.g. Jones 2007b showed an average compensation point of >0.4 µg m^{-3} for moorlands at background atmospheric concentrations). This is principally confirming that low concentrations in the range of the suggested critical levels can be "sensed" by vegetation in pristine areas, but as mentioned above, ammonia concentrations in agricultural and densely populated regions will only rarely be that low. During the lifetime and with the biological activity of plants during seasons, fluxes of ammonia will change. While young N-demanding plants are often a sink of ammonia in spring, old senescing plants may be a source and release the gas in the autumn. In addition, ammonia can be transported from a highly fertilized crop canopy to an adjacent natural vegetation with a lower compensation point and vegetation on acidic soil substrates will probably be more prone to the deposition of ammonia.

Following the initial discussion of critical levels for ammonia at the UNECE Workshop in Egham 1992 (Ashmore and Wilson 1994), European research networks had been established in the 1990s to monitor atmospheric processes that lead to the deposition of oxidized and reduced nitrogen species. It became clear that ammonia has a rather short life-time (Asman et al. 1998) reaching acute concentrations only close to emission sources. Further, chronic responses in sensitive wild plants will be driven by the joint deposition of various N-containing compounds mainly in the oligotrophic ecosystems. In order to account for the differences between short-term atmospheric concentrations and the cumulative annual inputs, the concept of critical loads had been introduced.

Ground-breaking research on the adverse effects and dose-response functions of ammonia on the natural (e.g. nitrogen-poor heathland) vegetation came from the Netherlands and served as the basis for the UNECE critical level of 8 µg m^{-3} (van der Eerden et al. 1982; Dueck 1990; van der Eerden et al. 1991; Dueck and Elderson 1992). Research of van der Eerden (1982) was based on the use of diluted exhaust from stables in closed or open top chambers (OTC), while later studies in Wageningen (Dueck and van der Eerden) involved technical ammonia supplied via mass flow controllers. Concentrations were in the range of 50 to 100 µg m^{-3} and were determined with NOx converters/monitors. While there were several OTC experiments in the UK addressing NH_3 and wet deposition of N in the 1990-2000s, the only comparable research project performed in Germany dealt with an ammonia fumigation of three grassland species grown in monocultures and mixtures. The experiments were performed in OTCs with diluted technical ammonia using set concentrations of 20 and 50 µg m^{-3} (Jäger et al. 1998). However, the realized concentrations could not be documented due to failures of the NOx converters/monitors. Summaries of the pioneering research of ammonia effects on plants can be found in the reviews of Adaros and Dämmgen (1994), Fangmeier et al. (1994) and Krupa (2003).

Based on the above-mentioned studies, we suggest that ammonia concentrations were probably too high in the chamber-based experiments from 1970 to ca 2000, restricting the validity of results with respect to the old and new critical levels. Expert judgements and extrapolations from high to low concentrations were used and long-term responses were predicted from short term experiments. None of the mentioned fumigation experiments operated in the range of existing environmental concentrations mainly due to technical reasons. When using pure ammonia without a time and concentration-controlled feedback system, peaks and accidental overdosing may occur that will lead to short-term burdens on plants eventually triggering acute effects. In fact, this will likely also happen in natural vegetation that is exposed to the fumes and plumes from adjacent slurry spreading or livestock housing.

Early Ammonia Research in growth chambers

- Focus on high concentrations and crop species, mainly in the Netherlands and the UK
- Mainly short-term experiments
- Pure (technical) NH_3 supply

Table 1: Overview of ammonia fumigation experiments in closed and open chambers

Source	Fumigation type	Supply of ammonia	Set range in µg m^{-3}	Method of ammonia measurement	Data on realized concentrations	Duration	Plant species
Faller (1972)	Glass chambers, 0.5 m^3	diluted NH_3, daylight hours	0 - 1600	none, calculated from flow	No	3 weeks	*Helianthus annuus*
Hutchinson et al. (1972)	Plexiglass chambers, 0.3 m high, 0.15 m sides	diluted NH_3,	24-44	trapped in KCl solutions	24-44	24 hrs	*Glycine max, Zea mays, Helianthus annuus*
Ewert (1979)	2 chambers of 10 m^3	diluted NH_3, 24 hrs day^{-1}	700 - 21000	air sampling, Nessler´s reagent	No	1-500 hrs	48 woody species, 10 crops
Farquhar et al. (1980)	Glass chamber, 0.2 m^3	diluted NH_3, 24 hrs day^{-1}	0 - 50 nbar	air sampling, indophenol method	No	20 minutes	*Phaseolus vulgaris* L. and other
Cowling & Lockyer (1981)	Exposure chambers	diluted NH_3, 16 hrs day^{-1}	0, 148 and 550	air sampling, Nessler´s reagent	No	40 days	*Lolium perenne* L.
van der Eerden (1982)	Open Top Chamber, closed chambers	Exhausts from poultry and pig manure, 24 hrs day^{-1}	600 - 4500	air sampling, Nessler´s reagent	No	3 - 60 days	grasses, trees, *Brassica*, *Lycopersicon*
Lockyer & Whitehead (1986)	Exposure chambers	diluted NH_3, 16 hrs day^{-1}	0, 100 and 500	air sampling, indophenol method	16, 118, 520	40 days	*Lolium multiflorum* Lam.
van Hove et al. (1987)	Leaf Chamber	diluted NH_3, short term	0 - 400	Chemiluminiscence after converting NH_3 to NO	No	some days	*Phaseolus vulgaris* L.
Whitehead & Lockyer (1987)	Exposure chambers	diluted NH_3	0 - 700	air sampling, indophenol method	14 - 709	33 days	*Lolium multiflorum* Lam.
Dueck (1990)	Open Top Chambers	diluted NH_3, 24 hrs day^{-1}	0, 53 and 105	Chemiluminiscence after converting NH_3 to NO	No	9 months	*Calluna vulgaris*
Dueck & Elderson (1992)	Open Top Chambers	diluted NH_3, 24 hrs day^{-1}	0, 50	Chemiluminiscence after converting NH_3 to NO	No	9 months	*Arnica montana, Viola canina, Agrostis capillaris*
Husted & Schjørring (1996)	Glass cuvette, 0.45 m^3	diluted NH_3, 24 hrs day^{-1}	0 - 25 nmol mol^{-1}	Chemiluminiscence after converting NH_3 to NO	No	twice a day, 7 days	*Brassica napus* L.
Jäger et al. (1998)	Open Top Chambers, 3.15 m diameter	diluted NH_3, 24 hrs day^{-1}	20 and 50	Chemiluminiscence after converting NH_3 to NO	No	3 years	Monocultures and mixtures of *Bromus erectus* Huds, *Brachypodium pinnatum* P. Beauv. and *Arrenatherum elatius* P. Beauv.
Geßler et al. (2002)	Borosilicate twig chambers	diluted NH_3, 24 hrs day^{-1}	100	continuous wet flow denuder AMANDA	No	several hours	*Picea abies*
Adrizal et al. (2006)	Exposure chambers	diluted NH_3, 24 hrs day^{-1}	2800 - 5000	Photoacoustic NH_3 detector	No	12 weeks	*Juniperus virginiana*, *Gleditsia triacanthos*, *Phalaris arundinacea*, *Populus* spec. *Salix* spec.

Table 1 continued

Source	Fumigation type	Supply of ammonia	Set range in µg m^{-3}	Method of ammonia measurement	Data on realized concentrations	Duration	Plant species
Jones et al. (2007a)	Flux chamber, 1.84 m^3, placed in an OTC at Bush Estate near Edinburgh, Scotland	1% aqueous NH3 solution at different rates and concentrations	1-100	AMANDA system (Ammonia Measurement by ANnular Denuder sampling with on line Analyser system)	daily resolution	6 hours each trial over 3 to 6 weeks	mixed turfs made up of *Calluna vulgaris* (L.) Hull, *Eriophorum vaginatum* L., *Sphagnum capillifolium* (Ehrh.) Hedw. and *Sphagnum papillosum* Linb.
Jones et al. (2007b)	Flux chamber, 1.84 m^3, placed inside of an OTC	1% aqueous NH_3 solution at different rates and concentrations	1-140	continuous wet flow denuder AMANDA, measurements at inlet and outlet	daily resolution	6 hours each trial over 3 to 6 weeks, stomatal and non-stomatal species	*Deschampsia cespitosa* (L.) Beauv., *Calluna vulgaris* (L.) Hull, *Eriophorum vaginatum* L., *Cladonia* spp., *Sphagnum* spp., and *Pleurozium schreberi* (Brid.)
Jones et al. (2008)	Flux chamber, 1.84 m^3	release of ^{15}N labelled NH_3 from $(^{15}NH_4)_2SO_4$ with NaOH	16 - 20	continuous wet flow denuder AMANDA	daily resolution	15 days	*Calluna vulgaris*
Franzaring et al. (2012)	Mini greenhouses	diluted NH_3, 24 hrs day^{-1}	0 - 200	Passive samplers	2.2 -195	88 days	*Brassica napus* L.
Ilogu Chibuzo (2012)	Exposure chambers	controlled evaoporation of NH_4Cl solutions	0 - 84	Passive samplers	3, 25, 84	30 days	*Echinochloa crus-gallii* and *Lolium multiflorum*
Jones et al. (2013), presenting results from PhD Thesis of Jonathon Foot from 1998	Open Top Chambers	uncontrolled evaporation of NH_4OH solutions containing 0-10 % of ammonia	0 - 35	Passive samplers	Yes	28 weeks	Mixtures of *Dactylis glomerata, Plantago lanceolata, Festuca rubra, Centaurea nigra* etc.

2.2 Research after 2009

It must be highlighted that during the last 30 years many research activities on the fluxes, deposition and effects of ammonia came from Scotland, namely David Fowler, Neil Cape and Mark Sutton, all scientists from the Centre for Ecology & Hydrology (CEH) Edinburg. These authors have co-authored many studies and worked as PI´s of several EU projects that dealt with the deposition of acidifying and eutrophying gases and particulates. We only cited a few of these studies in previous chapter, but acknowledge the impact they still have on current research.

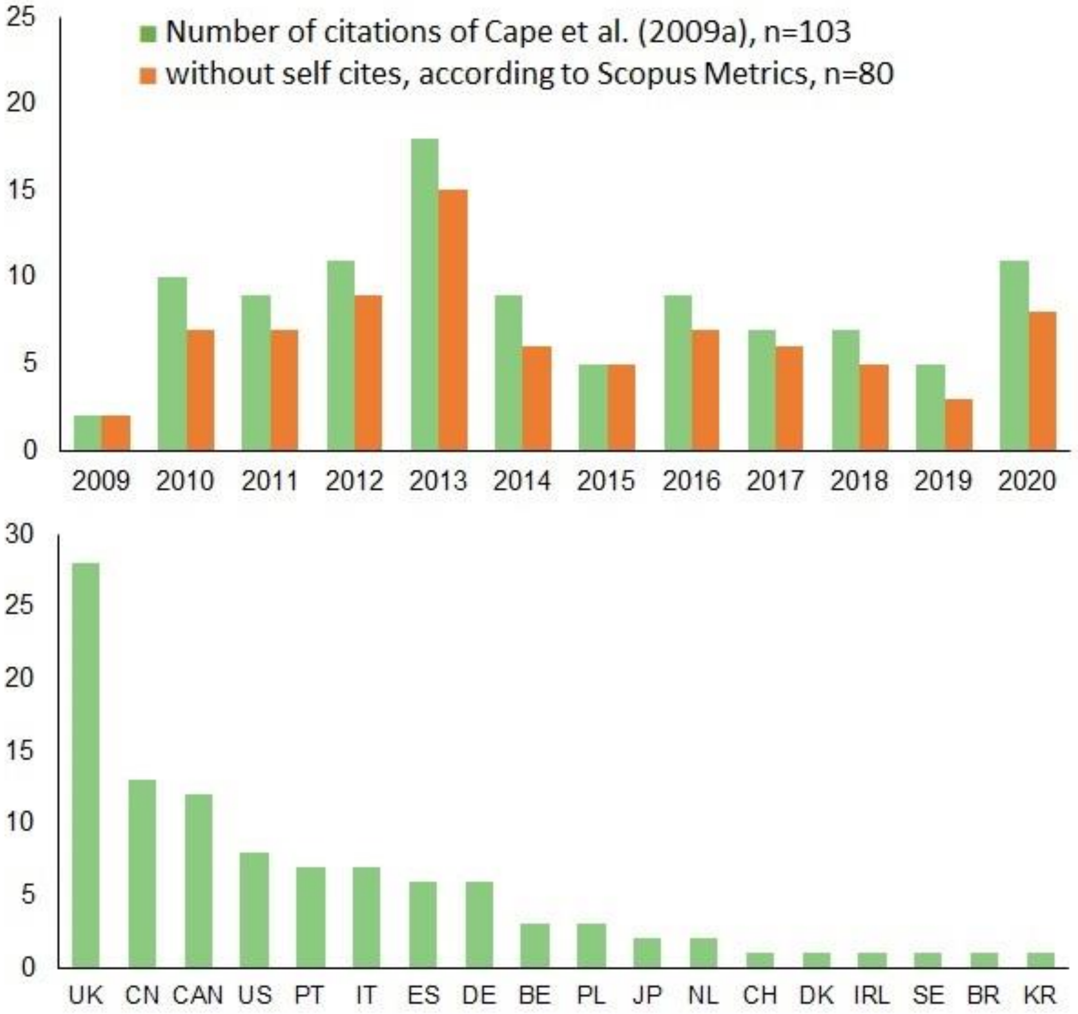
In present sub-chapter we will take a closer look at research that deals with the effects of plants and the vegetation under more natural conditions. Whereas chamber-based experiments (see. 2.1) dealing with plant exposure to ammonia were mostly applying higher concentrations, more recent projects tried to investigate low-range ammonia effects in order to address potential responses in sensitive plant species and to be more representative for the range of critical levels.

A screening of newer literature dealing with the recommended CLe and contributing new data from the UK and other European countries will be presented in this subchapter. We present an overview of 103 peer-reviewed references (as of December 2020) that cited Cape et al. 2009 according to the databank Scopus®. Figure 1 (top) gives an overview on how many citations per year were recorded and how many of these did not stem from the Scottish research group that also authored the Cape et al. publication. After a decline in the numbers of citations until 2019, the topic "critical levels" peaked again recently and we can observe that 80 % of the citations were from groups not involved in writing the cited paper.

When looking at the country of residence of the main author and her affiliation (Figure 1 below), we can see that most publications which cited Cape at al. (2009) stemmed from the UK, China, Canada and the US, being followed by three Mediterranean countries mostly studying lichen diversity among ammonia gradients. Interestingly, in the Netherlands, Denmark and Germany, the research on effects of ammonia is not of major importance, although the countries have a very large livestock production sector and severe problems with eutrophication. It must be noted that until the late 1990s, most of the ground-breaking research on ammonia impacts had been performed in the Netherlands (the research group of Dr. Ludger van der Eerden at Plant Research International in Wageningen). The old CLe of 8 µg m^{-3} had originally been derived from fumigation studies from that institute and in those years, there were many ammonia-related research programs funded by the Dutch, UK and German governments. An even stronger absence of research was noted in France, a country with a strong livestock sector in the EU, where no peer-reviewed literature on the effects of ammonia was published after 2009.

When looking at the specific topics dealt with in the 103 articles, the study of the presumed ammonia sensitivity of lichens plays an overwhelming role. 27 articles focus on the mapping of nitrogen sensitive lichens with many published from Southern European countries, despite the limited intensive livestock. Another quarter of the citing articles dealt with the modelling of N deposition and 20 articles were presenting data from ammonia measurements using passive samplers. 16 studies investigated changes in the species composition. 11 studies reported about "bioindication" methods, addressing ecophysiological changes in lower and higher plants along ammonia gradients. Only a few of these studies used pre-cultivated higher plants, which were actively exposed in the field at locations, where ammonia was measured. In the recent paper of Ellis et al. (2022) on potential nitrogen risks to Himalayan forests, the results of 14 lichen biomonitoring studies were used to derive independent estimates for three nitrogen pollutant types. These were the critical level for ammonia concentration (µg m^{-3}), the critical load for total nitrogen deposition (kg N ha^{1} yr^{-1}) and the total throughfall nitrogen deposition. Based on their calculations for diverse responses on lichens (e.g. total species richness, sensitive species richness, 'forage lichen' abundance and cyanolichen abundance), the critical level for ammonia would be 1.44 µg m^{-3} as a grand mean and the critical load (wet and dry, reduced and oxidized) would average to 8.26 kg N ha^{-1} yr^{-1}.

Figure 1: Citations of Cape et al. (2009) in the peer-reviewed literature, broken down after years (top) and countries of residence of the main author (below)



Source: Based on a literature search in the SCOPUS database © Elsevier B.V

Besides the agricultural sources, the NOx emissions from industry, traffic and wastewater treatment sectors are also responsible for the losses in biodiversity of certain cryptogams and lichens. Sutton et al. (2020) have recently suggested that the successful reduction of SO_2 pollution in Europe and elsewhere has led to a general alkalinisation of rain and air that will be harmful to naturally acidophytic species. On the other hand, in the 1990s and early 2000s the elimination of acidification often favoured the return of rare basiophytic lichens. The impacts of the recent and ongoing changes in air quality have also been investigated in Sweden and showed a very slow recolonization of sensitive species due to existing dispersal limits (Weldon and Grandin 2021).

Only in the 1990s, research on the phytotoxicity of air pollutants moved to field-based fumigation systems to study the effects of ammonia on pre-cultivated plants or established natural vegetation. Such systems should operate under the normal ambient climates and should not create concentration peaks. Examples are the FACE (free air carbon dioxide enrichment) experiments and the use of open top chambers (OTC) e.g. to study adverse effects of ozone.

The only long-term field ammonia fumigation system is at the Scottish Whim Bog Field Facility (Southwest of Edinburgh) and has been in operation since 2002. The site simulates a point source release (e.g. chicken farm) producing a concentration gradient across an ombrotrophic bog, ranging from an annual average of around 80 µg m^{-3} (at 6 m) to ambient (<1 µg m^{-3}). The site represents a release of approximately 5.000 chickens in a clean shed system (SCAIL model estimate) and is similar to annual concentrations reported near comparable farms (e.g. Pitcairn 1998). All major species have declined (including *Cladonia, Sphagnum, Calluna, Hypnum*) except the now dominant sedge *Eriophoum vaginatum*, across the large majority of the concentration ranges (van Dijk et al. 2017; Levy et al. 2019). Evidence suggests long term effects with impacts responding to changes occurring over several years (Sutton et al. 2020). Severe impacts of lichen bleaching recorded near the source may be caused by peak toxicity events, but impacts along the transect are likely caused by other mechanisms such as algal/fungal imbalances.

More recently in 2021, CEH has established a new field ammonia fumigation system at Glencorse Field Facility (Southwest of Edinburgh). This site releases ammonia at a similar level to Whim, but assesses the impact of ammonia within a woodland environment (pers. comm. Matthew Jones). The site has 20 m long fumigation pipes at three heights to ensure ammonia is relatively well mixed in the trunk region of the woodland. Ammonia is only released when wind directions and speed are adequate to ensure maximum concentrations are representative of concentrations expected downwind of intensive agriculture. Significant pre-testing of the system was conducted using a Picarro gas concentration NH_3 analyser in a multi-height inlet system, multi-height profiles of ALPHA samplers, a transect of ALPHAs and ALPHAs placed throughout the woodland to allow the researchers to establish the plume shape and concentrations under different meteorological conditions. The site also has a 15 m high meteorological mast to establish wind, temperature, relative humidity and leaf wetness profiles. The site is being used as a test bed to improve – model estimates of NH_3 deposition to woodlands, NH_3 impacts on forest ecosystems, NH_3 impacts on GHG emissions, and the work will include assessment of NH_3 impacts on lichen transplants in relation to critical levels. A third site has also now been established (March 2022) in Sri Lanka following the same intensive pre-measurements as at the Glencorse experiment, which will for the first time assess nitrogen impacts in South Asian ecosystems, and will be compared to a large-scale assessment of N impacts in the Himalayan Forests in South Asia.

A new generation of experiments mimicking the dry gaseous deposition of ammonia in established ecosystems is currently on the way also in China (Pan et al. 2020) and will be recognized under the name Free-Air NH_3 Enrichment (FANE).

Critical Levels Research

- Until 2020, Cape et al (2009) had been cited over 100 times
- Most citations stem from UK, China, Canada and the US
- Relevant number of publications in several Mediterranean countries
- Low number of publications from the Netherlands, Denmark, Germany and France, although the countries have a large livestock sector and eutrophication problems are common
- Citing studies deal with lichen mapping, modelling N deposition, ammonia measuring, changes in species composition and the investigation of ecophysiological changes in lower species
- Critical loads research is more prominent than the study of ammonia or nitrogen dioxide

2.2.1 Gradient Studies

Most field experiments dealing with the effect of ammonia used passive approaches working with the reaction of established vegetation on ammonia input. Sheppard et al. (2011) described a dramatic loss of higher and lower plants at Whim Bog after three years of relatively modest NH_3 deposition, with impacts after 14 years of treatment being observed down to low NH_3 concentrations (Levy et al. 2019). Several lichen mapping studies also showed effects of ammonia with regard to lichen diversity along natural gradients. The number of oligotrophic lichen species decreased significantly with increasing atmospheric ammonia concentrations whereas nitrophytic species increased and/or replaced some species (Pinho et al. 2011; Pinho et al. 2012; Pinho et al. 2014). Pinho et al. suggested CLe of ammonia for Mediterranean evergreen woodlands to be 1.9 µg m^{-3} (2012) and two years later they suggested a lower level of 0.69 µg m^{-3}. Aguillaume et al. (2017) suggested a higher CLe of 2.6 µg m^{-3} for a Mediterranean Holm-Oak Forest in Spain, but argue that the site has already been polluted by NH_3 in the past. Another study from Spain monitored NH_3 in peri-urban forests (*Quercus ilex*) and found no CLe exceedance in any of the three sites (García-Gómez et al. 2016). It was furthermore shown, that below-canopy concentrations of NH_3 were around 40% lower than in open-field plots, indicating a high sink capacity of holm oak forests.

Mayer et al. (2013) showed that lichen diversity in Austria has been declining in a remote area (in the European Alps) and suggest eutrophication due to ammonium in precipitation and fog as an important factor. Experiments from Canada revealed that most lichen species studied were affected above 1.5 µg m^{-3} (Watmough et al. 2014). Izquieta-Rojano et al. (2018) describe effects of an ammonia gradient (distance to a swine livestock facility) on mosses (especially tissue N content) in an oak woodland with an estimated site-specific CLe of 3.5 µg m^{-3}. The results imply that mosses might stand higher concentrations of ammonia than lichens, but it must be noted that there is less information on mosses compared to lichens at the moment. Other passive approaches like the use of oak leaf characteristics as a biomonitoring tool. (Wuytack et al. 2013) revealed no differences and therefore seem not to be suitable for ammonia monitoring programs.

Active (biomonitoring) studies are mainly addressing the effects of ammonia from agricultural sites on lower plant species, i.e. lichen and/or moss diversity and vitality. Sensitive moss (Freiberger et al. 2021) or lichen species like *Evernia prunastri* (Paoli et al. 2010; Paoli et al. 2014; Munzi et al. 2014) or *Xanthoria parietina* and *Flavoparmelia caperata* (Frati et al. 2007) had been transplanted from non-polluted sites (sites with low ammonia background concentrations) to natural ammonia gradients and the vitality e.g. in form of chlorophyll fluorescence had been determined. Munzi et al. (2014) found that ammonia concentrations above 3 µg m^{-3} would abruptly decrease plant fitness and frequency but the authors approve the same NH_3 CLe (1 µg m^{-3}) as proposed by Cape et al (2009) because of possible cumulative effects after longer exposure than tested. Interestingly, a more recent study observed that the N-sensitive lichen *Cladonia portentosa* did not show any adverse effects anymore after ceasing the wet deposition of nitrogen (Munzi et al. 2020).

The effects of a higher availability of nitrogen along gradients do not only depend on the form of N and the wet/dry or gaseous/particulate deposition but are also affected by environmental gradients. Morillas et al. (2021) studied detrimental cumulative effects of solar radiation and increased nitrogen treatments on lichens via immersing samples into N solutions. Studies about ammonia effects are rarely including additional climatic factors to date. Freiberger et al. (2021) recently suggested that active approaches using uncontaminated moss transplants (*Pleurozium schreberi*) and accumulated N-levels show a much stronger relationship to the ammonia concentrations than moss or lichen mapping (passive approaches).

Gradient studies in the vicinity of specific emitters making use of higher plants are often focussing on grass species as bioindicators since their upright growth habit and the presence of numerous stalks enable the effective scavenging and accumulation of air pollutants. While Leith et al. (2009) used grasses in the lee of Scottish poultry farms (confirming CLe of 2-3 µg m^{-3}), Ilogu Chibuzo (2012) used grasses and N-demanding weeds in the lee of a pig stable in Southern Germany. In both studies, plant biomass increments and nitrogen concentrations proved to be sensitive indicators for the fertilizing effect of ammonia.

Gradient Studies

- Most field studies make use of passive approaches, i.e. investigate the reaction of established vegetation on ammonia concentrations in a natural gradient
- Many studies show a decrease of N-sensitive lichen species with increasing NH_3 concentrations
- Mosses seem to be more tolerant than lichens
- Most active study approaches with lichen transplants confirm low CLe
- Most recent studies addressing CLe for ammonia work on passive approaches with lichens in natural ammonia gradients, the majority suggesting CLe of 0.69-3.5 µg m^{-3}
- Only few studies suggest that lichens might stand higher NH_3 concentrations than expected
- Only very few studies address higher plants
- Active approaches confirm CLe of 1 µg m^{-3} for lichens and 2-3 µg m^{-3} for higher plants (e.g. grasses)

2.2.2 Fumigation Studies

Apart from the free-air exposure at Whim Bog, which uses the release of technical ammonia along a wind gradient, only two peer-reviewed papers on ammonia fumigation could be found that had been published after 2009. In our own experiment (Franzaring et al. 2012), we studied the effects of sub-acute concentrations of ammonia on the re-growth of winter oilseed rape. We could show that the crop is well able to benefit from the gaseous fertilizer at a concentration of 195 µg m^{-3}, but we did not test whether slightly raised or low concentrations of ammonia would be able to significantly interact with the physiology of the species.
The second study on ammonia fumigation was published by Jones et al. (2013). The authors investigated the effects of ammonia deposition to plants from Welsh sand dune ecosystems and exposed mesocosms of plant mixtures in OTCs. Shoot biomass and tissue nitrogen increased significantly with increasing concentrations of ammonia. A significant biomass increase of the plant mixture was already observed when increasing the ammonia concentrations from 0.4 to 4.3 µg m^{-3}, i.e. dose equivalents of 2 and 24 kg N ha^{-1} y^{-1}.

Fumigation Studies

- Apart from the Whim Bog Experiment, only two newer fumigation studies were found
- Both studies showed a biomass increase of the species tested under NH_3 fumigation

2.3 Ammonia monitoring

Since the 1990s, pioneering work on ammonia measurements in the ambient air has been done in the Dutch Landelijk Meetnet Luchtkwaliteit (LML) and in the UK National Ammonia Monitoring Network (NAMN). Various types of denuders and passive sampler devices are being applied to allow for a resolution ranging from days to weeks.

More recently, other devices have been made available for fast, continuous, real-time monitoring of ammonia. Examples are devices based on Differential Optical Absorption Spectroscopy (DOAS), Cavity Ring-Down Spectroscopy (CRDS) or Quantum Cascade Laser Absorption Spectroscopy (QC-LAS) While those methods are well able to determine low background concentrations, they are typically not employed in monitoring networks due to their high cost and high demand for support. In the Netherlands and in Switzerland a few monitoring sites employ DOAS devices (Volten et al. 2012a; Volten et al. 2012b; Berkhout et al. 2017; Bell et al. 2017) and more CRDS instruments may be included in monitoring initiatives in the near future (see Pogány et al., Poulain and van Zwieten, in this issue).

A major challenge in measuring ammonia concentrations is its stickiness to surfaces of measurement devices. Therefore, there is a high need for more intensive metrological research focusing on the quality assurance, inter-comparability and validation of ammonia in the ppb range.

A first international initiative comparing different active and passive analysers was set up within the European MetNH3 (Metrology for ammonia in ambient air) project. One of the work packages of the MetNH3-Project dealt with the testing and calibrating of ammonia passive samplers. Samplers were tested in the UK's National Physical Laboratory's (NPL) controlled atmosphere test facility (CATFAC) and in the field at the above-mentioned Scottish Whim Bog Experimental site. The inter-comparisons between different sampler types have been presented by Braban et al. (2018) and Martin et al. (2019). Measurement errors of the tested passive samplers varied between 11 and 23 % at the reference concentration of 1 µg m^{-3}, i.e. a concentration representative for the lower critical level. ALPHA passive samplers proved to be the best suited for the lower concentration ranges and have an exposure time of 28 days, whereas e.g. the Radiello® samplers can be used to investigate shorter periods.

Most recently, a comprehensive comparison of measurement techniques under field conditions has been presented by Twigg et al. (2022) and will be discussed by the research community.

Here we focus mainly on the results determined by passive samplers. These cost-effective devices have been developed for the determination of gaseous air pollutants in locations without power supply and are therefore the preferred method to monitor gas concentrations in remote areas. In order to assure their quality, devices should be certified according to CEN (European Committee for Standardization) standards EN 17346 (2020)[1] or comparable national standards for diffusive samplers. Two types are available, axial (badge and tube types) and radial (tube like) samplers. Latter types have higher sampling rates and can therefore be exposed over shorter times (Puchalski et al. 2011).

Table 2 gives an overview of studies that used passive samplers to study ammonia concentrations in different countries. Only papers and grey literature are listed that were published after 2009. In most of the studies, ammonia concentrations did not decrease over time contradicting the results from modelled concentrations (Wichink Kruit et al. 2017) and the information derived from emission inventories. As previously mentioned, the reasons for higher levels may be the reduced concentrations of NO_x and SO_2 and increased volatilization of the gas due to the higher ambient

[1] https://www.en-standard.eu/csn-en-17346-ambient-air-standard-method-for-the-determination-of-the-concentration-of-ammonia-using-diffusive-samplers/

temperatures. At the same time, higher concentrations may also stem from previously underestimated emissions, e.g. due to uncertainties in emission factors or activity data in agriculture, from waste water treatment facilities and biogas plants as well as from the increased use of diesel exhaust fluids (DEF, AdBlue). Higher vehicular emissions of NH_3 and ammonia slip have been addressed by Elser et al. (2018), Suarez-Bertoa and Astorga (2016) and Fenn et al. (2018).

In contrast to the pioneering, smaller European countries like the Netherlands (Lolkema et al. 2015; Noordijk et al. 2020), Belgium (VMM 2017) and Switzerland (Thöni et al. 2018; Seitler & Meier 2022) and the UK (Tang et al. 2018), countries with intensive agricultural systems like France, Italy and Germany do not operate national ammonia networks yet, so that regional data are scarce in these countries. We could only find one publication from France that reports on ammonia measurements in five agricultural and six forest regions (Fauvel et al. 2019). In Brittany, a region with intensive livestock farming, ammonia concentrations are on average in the range of 9 µg m^{-3}, while inside forest areas in the south and in the east, concentrations of less than 1 µg m^{-3} are common. In Germany, there have been some efforts in various federal states to monitor ammonia with passive samplers, mainly badge type devices (e.g. Lohrengel et al. 2012; LfU 2019).

Belgium, the Netherlands and Ireland have established monitoring sites in a large number of Natura2000 (the EU Habitat Directive) areas (Table 2). In contrast, in Germany measurements are being performed on a voluntary basis and are not conducted with uniform objectives in eleven out of sixteen Federal States. Since protected landscapes are often nitrogen limited, the vegetation (e.g. bogs and heathlands) is an important receptor for potentially harmful ammonia. In the present workshop proceedings, results from some European countries and Northern America will be presented in greater detail (see the contributions of Staelens, Meier, Kelleghan, Gay & Puchalski, Moravek & Geupel, this issue).

Ammonia Measuring and Monitoring

- Ammonia monitoring programmes have been set up in both, N-America and Europe. Also, in China, NH_3 is being determined at many locations of the national N deposition networks.
- The intensity and technology used for ammonia monitoring differs between countries. The Netherlands and Switzerland are routinely operating a few sites with high-precision trace gas measurements, while in other countries DOAS and CRDS are applied in short-term research projects only.
- Many countries and studies use denuders and passive samplers, latter method representing a well-accepted, feasible low-cost technique to measure NH_3.
- Real-time monitoring gas analysers are mainly used in temporary, small-scale initiatives. Their continuous development towards lower maintenance requirements and easier handling may make them more suitable for continuous monitoring of ambient levels of ammonia in the future.

Table 2: Overview of ammonia monitoring programs

List is based on knowledge of monitoring programs to authors at the time of publication and may be incomplete.

Country	Sampler type	Network name	N of sites[2]	Period	Network Details / Results	Sources
BE	Radiello®, blue diffusion barriers, miniDOAS		100 Natura2000 sites, miniDOAS at 2 sites	2015-2016	Country mean: 3.3 µg m^{-3} NH_3, in Western Flanders > 5 µg m^{-3} NH_3, no trends over time.	VMM (2017)
CAN-ONT	Ogawa	NAPS	74	2006-2007	Even in remote sites in CAN, 50% of the sites would exceed the critical levels.	Yao & Zhang (2013; 2019)
CH	Radiello®		13	since 2000	No reductions over time, different levels in different regions. All values exceed the 1 µg level, most the 3 µg critical level.	Seitler et al. (2018); Seitler & Meier (2022)
DE -Depo	FERM (IVL)		15 forest sites (in 2020)	since 2001	ICP-Forests Level II locations, bi-weekly. No decline in concentrations in German Forests. Grand mean: 1.9 µg m^{-3}.	Thünen Institute of Forest Ecosystems (pers. comm.)
DE-BW	FERM (IVL)		16[3]	since 2007	Selected urban and rural locations. Moderately high NH_3 at traffic sites.	LUBW (https://pd.lubw.de/10334)
DE-BY	Ferm (IVL)	LÜB, DBS	15	since 2011	Selected locations, four-weekly. Downward trend in cities, in rural areas increasing trend since 2017. Yearly mean levels up to 3 µg m^{-3} at rural sites, 7 µg m^{-3} at intensive agricultural sites.	LfU (2019) and LfU (pers. comm.)
DE-MV			10	2010	No trend study possible.	https://www.lung.mv-regierung.de/umwelt/luft/archiv/jb_2019_small.pdf
DE-NRW	Ferm (IVL)		5	since 2001	Selected urban and rural locations. No trend.	Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen (pers. comm.)
DE-NI	Ferm (IVL)		19	since 2010	Selected rural locations. 0.7 to 13 µg m^{-3} ammonia. Two sites in industrial settings.	https://www.umwelt.niedersachsen.de/download/183444/Jahresbericht_2021.pdf
DE-SH			10	2008	No trend study possible.	https://www.schleswig-holstein.de/DE/fachinhalte/L/luftqualitaet/Downloads/Berichte/bericht_orient_messungen_NH3_SH_2011.pdf?__blob=publicationFile&v=1

[2] If measurements are still ongoing, number represents the current number of sites with operating ammonia measurements
[3] BW established 13 new sampling sites in 2021. They are not officially documented, therefore not reported here

Country	Sampler type	Network name	N of sites	Period	Results	Sources
DE-ST			7	since 2021/22	No trend study possible.	LUA (https://lau.sachsen-anhalt.de/luft-klima-laerm/lufthygienisches-ueberwachungssystem-luesa/)
DE-UBA	Denuder		7	2004	Network background sites of the German Environmental Agency	UBA (https://www.umweltbundesamt.de/das-uba/standorte-gebaeude#dessau)
F	Alpha, Delta		11	2009-2019	Selected agricultural and forest locations. High ammonia concentrations in Brittony.	Fauvel et al. (2019)
IRL	Alpha		12 Natura2000 sites	2017-2018	0.47–4.59 µg NH_3 m^{-3}. Agriculture is the main source and ammonia is related to ambient temperatures.	Kelleghan et al. (2021)
LUX	Gradko		4	since 2019	Mean concentrations vary from 5.5 to 19.2 µg m^{-3} at rural and agricultural sites.	Pers. comm. Trebs & Junk, Luxembourg Institute of Science and Technology (LIST 2022)
NL	Gradko, miniDOAS	MAN	Currently 310 measuring points at 105 sites, out of which 77 are Natura2000 sites, mini DOAS operated at 6 sites	since 2005	NH_3 concentrations significantly increased in easterly regions and went down in the west and in the south.	Lolkema et al. (2015); Noordijk et al. (2020); pers. comm. Jonkers and Bleeker
UK	Alpha, Delta	UK NAMN	74	since 1998	Reduction of 3% on national level, but ammonia increases in some regions.	Tang et al. (2018); United Kingdom Eutrophying & Acidifying Network (UKEAP), https://uk-air.defra.gov.uk/networks/
US	Radiello®, blue diffusion barriers	AMoN	50	since 2007	NH_3 concentrations have increased by 7% between 2008 and 2015 at 18 long-term sites.	Puchalski et al. (2011) and Yao & Zhang (2016; 2019)

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3 Discussion points at the workshop

Before and during the workshop, we aimed to collate recent information on whether critical levels of ammonia need to be revised and asked the presenters and participants to notify the audience about ongoing research activities. In order to prepare a general discussion, we raised the following questions prior to the workshop. Participants were asked to respond before the meeting, but we did not get any feedback until the workshop. However, we were still able to address some of the points at the final discussion (see chapter 4.3)

Discussion 1: Current set-up and future perspective on ammonia monitoring

- With regard to ammonia measuring, is the use of passive samplers suitable? At which frequency and at how many sites should samplers be analysed?
- Are there new/suitable/affordable measuring devices to continuously monitor NH_3 in the low concentration range?
- Which criteria have to be met to set up a national representative ammonia network?
- Which difficulties complicate ammonia monitoring networks? Costs?
- Is there essential knowledge for recommendations to policy makers that is gained (exclusively) with ammonia monitoring networks?
- Is ammonia monitoring beneficial for nature protection and air quality policies?
- Would you support an ammonia monitoring obligation and for what reasons?

Discussion 2: New findings with potential relevance to NH_3 CLe: Future directions

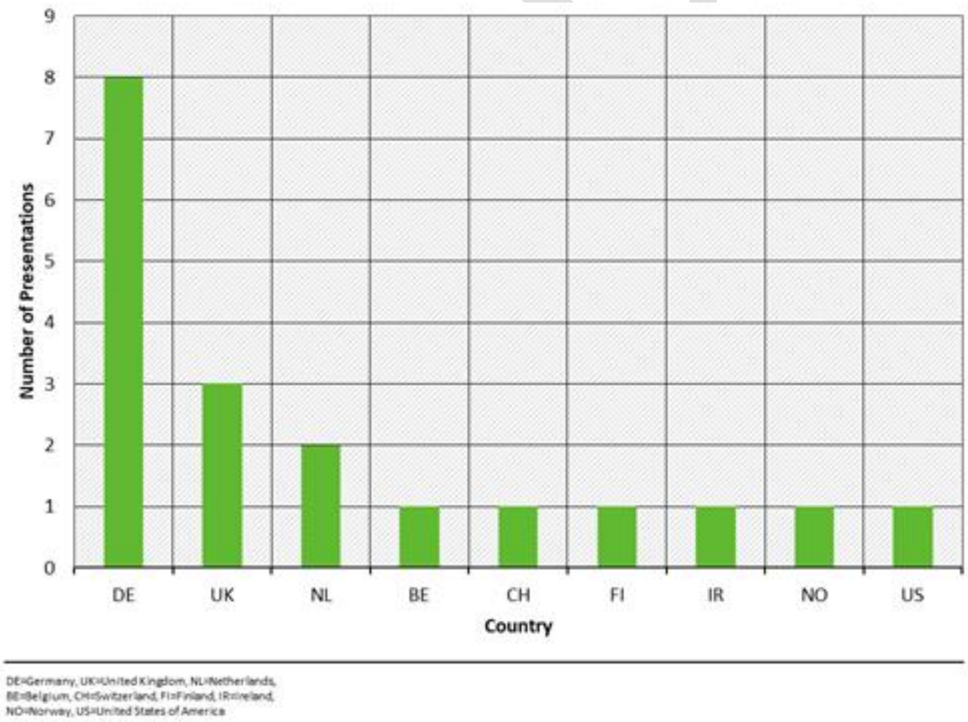
- Do you perform or are you aware of research on ammonia effects on higher or lower plants using controlled fumigation at ammonia levels < 10 µg m^{-3}? What are the technologies involved?
- Do you perform or do you know of ammonia fumigation studies in the field? What are the technologies involved and which are the tested ecosystems?
- Do you perform or do you know of gradient studies using active or passive biomonitoring, i.e. the use of vegetation on-site or plants being actively exposed? What are the involved technologies and plant species?
- Would you support annual, monthly and/or short-term CLe?
- Would you support to include knowledge on Critical Loads into scientific justification of critical levels?
- Based on current research, is there (enough) evidence for changing/keeping the CLe (for lower and higher plants, respectively)?
- Which further steps after the Workshop should be taken in the review of NH_3 CLe?

4 Workshop presentations and discussion

The workshop on CLe for ammonia took place as a hybrid meeting on 28/29 March 2022 with the option of physical attendance at the German Environment Agency in Dessau, Germany. 15 participants were able to attend in presence and over 100 people were following the live stream via a WebEx online meeting platform from several European countries and the US. In total, there were 145 registered participants from 28 countries including representatives from several bodies to the convention such as the UNECE Secretariat, WGE, ICP Vegetation, ICP Forest, CDM, CIAM, MSC-West, TFRN and representatives from several EU Environment Agencies.19 presentations from presenters from nine countries (Figure 2) were dealing with a current review, models and future trends of NH_3 across Europe in the first session, different ammonia monitoring networks in the second session and with vegetation effects (recent research on different scales) in the third session.

The workshop started with a welcome message from Markus Geupel (UBA) introducing the aim and ideas of the workshop. As the last ammonia workshop took place in 2006 in Edinburgh, the first presentation dealt with a recap of the issues and topics back then. Then, a presentation about recent and first research on ammonia effects on vegetation was given, summarizing the most important points of a background document (updated version in Chapter 2 or present report). The rest of the presentations on day 1 addressed modelling and monitoring approaches in different countries.

Figure 2: Numbers of presentations per country



Source: own illustration

In the following, we present the workshop agenda and in the sub-chapters thereafter, we collate the written summaries which were provided by the presenters. Finally, we document the most important discussion points and come up with the key findings.

Expert Workshop on Ammonia

28.-29. March 2022

Held hybrid: German Environment Agency, Dessau / webex

All times are given in CEST

More than ten years after the recommendation of updated CLRTAP critical levels for ammonia (Mapping Manual), new findings on the effects of ammonia on vegetation have been discussed on a workshop prepared by CCE and Germany. Furthermore, the workshop offered the opportunity to exchange information on national or regional programs which have been set up for the monitoring of ammonia in sensitive habitats.

Scientists dealing with research on effects of ammonia on vegetation and ecosystems or monitoring ammonia in the environment were encouraged to present their research.

The expert workshop was held on 28 to 29March 2022 as a hybrid meeting. The option for a physical attendance was in Dessau, Germany, where the German Environment Agency (UBA) is situated. In parallel, online participation was offered. The workshop was part of the current work plan for the year 2022-2023 of CLRTAP (1.1.1.22) and was organized in the framework of an R&D Project in Germany financed be the German Environment Agency (UBA).

FINAL AGENDA (Number of presentations on day 1 is the same as in the sub-chapters)

All presentations can be accessed under https://clous.uba.de/index.php/s/Hk15ZLnlpF1ZWqb with the following access code: Ammoniak@uba

Day 1 (Monday 28 March 2022)

12.30-13.50 Session 1: Introduction (Background document, Current Review, models and future trends of NH_3 Concentrations across Europe)

Introduction to the workshop and the review process (Geupel, UBA, Germany)

1. **15 years after: Rationale and approach used in the 2006 Edinburgh review workshop (Sutton, CEH, United Kingdom)**
2. **Literature review on the effects of ammonia (Franzaring and Kösler, University of Hohenheim, Germany)**
3. **Modelling ammonia concentrations, trends and scenarios in Europe (Fagerli, MSC-West, Norway)**

13.50 - 15.10 Session 2: Ammonia monitoring networks (national, regional, in conservation areas)

4. **NH_3 concentration measurements (Bleeker, RIVM, The Netherlands)**
5. **Real-time monitoring of ammonia concentrations with Cavity Ring-Down Spectroscopy' (Hofmann, Picarro, The Netherlands)**
6. **Ammonia monitoring at long-term and temporary sites in Flanders (Staelens, Flanders Environment Agency, Belgium)**
7. **Monitoring atmospheric ammonia on Natura 2000 sites in the Republic of Ireland (Kelleghan, University College Dublin, Ireland)**

15.10 -15.40 Break

15.40 - 17.30 Session 2 continued

8. **UK Ammonia Network: present and future (Aazem, JNCC, United Kingdom)**
9. **Ammonia concentration measurements in Germany - an overview (Moravek & Geupel, UBA, Germany)**
10. **Metrological aspects to support environmental ammonia monitoring (Pogany, PTB, Germany)**
11. **Long-term NH_3 measurements at the TROPOS research station Melpitz using a MARGA (Poulain, TROPOS, Germany)**
12. **Long-term measurements to support deposition and critical loads assessments in the US (Gay, University of Wisconsin Madison & Puchalski, EPA, USA)**

17.30-18.00 Discussion 1: Current set-up and future perspectives on ammonia monitoring

Day 2 (Tuesday 29 March 2022)

9.00 – 10.40 Session 3: Recent research on different scales (fumigation chamber, field exposure, gradient studies with bioindicators, transplants and vegetation on site)

13. **Relationships between ambient NH_3 concentrations and epiphytic lichens in an urban environment (Manninen, University of Finland, Finland)**
14. **Results from an ammonia fumigation and a field study using endangered nitrogen-sensitive plant species (Kösler and Franzaring University of Hohenheim, Germany)**
15. **Towards understanding shorter-term critical levels with monthly resolution measurements (Dragosits, CEH, United Kindgom)**
16. **Biomonitoring of ammonia with the epiphytic lichen Hypogymnia physodes (Mohr, LWK Lower Saxony, Germany)**
17. **How can we assess the impact of project contributions of ammonia on Natura 2000 sites? (Uhl, FÖA, Germany)**

10.40 – 11.00 Coffee Break

11.00-11.40 Session 4: Interrelations of critical levels and critical loads

18. **Development of a trophic assessment and management system for habitat types (Directive 92/43/EWG) considering interrelations between critical levels and critical loads (Prüeß, LUBW Baden-Württemberg, Germany)**
19. **Ammonia concentrations in Switzerland compared to critical levels and loads (Meier, BAFU, Switzerland)**

11.40 - 13.00 Discussion 2: New findings with potential relevance to NH_3 critical levels: Future directions / next steps

4.1 Workshop Day 1, 28 March 2022

4.1.1 15 years on: Rationale and reflection on the 2006 Edinburgh Ammonia Workshop in the light of emerging evidence.

Mark A. Sutton[1], Patricia A. Wolseley[2], Netty van Dijk[1], Ajinkya G. Deshpande[1], Ulrike Dragosits[1], Marsailidh Twigg[1], Y. Sim Tang[1], Christine Braban[1], Pete Levy[1], William J. Bealey[1], Gothamie Weerakoon[2], Jize Jiang[3], David S. Stevenson[3], Buddhika Weerakoon[4], Sarath P. Nissanka[4] and Matthew R. Jones[1]

[1] UK Centre for Ecology & Hydrology, United Kingdom
[2] Natural History Museum, London
[3] University of Edinburgh, School of GeoSciences
[4] University of Peradeniya, Sri Lanka.

Background

A critical level for ammonia was first set in the 1980s at Bad Harzburg (Posthumus, 1988), which was subsequently reassessed in the 1992 Egham Workshop (Ashmore and Wilson, 1994). However, there were several arguments subsequently articulated for why further revision was needed. The original critical level included an annual value of 8 µg m^{-3}, but comparison with dry deposition rates showed that there was an inconsistency between critical loads and critical levels, with critical loads exceeded at much lower ammonia concentrations than critical levels. In addition, NH3 monitoring data suggested that there was an increased likelihood of exceedance of the critical level for averaging periods of longer than one year (see further below, regarding Figure 3), while field data suggested responses to NH3 concentrations in the field at much less than 8 µg m^{-3} (Sutton et al., 2009a). It was with these issues in mind, that the UK government agreed to sponsor the 2006 Edinburgh Atmospheric Ammonia Workshop (CEH, 2006), which led to revision of the ammonia critical level (UNECE, 2007) and a comprehensive publication that also covered other aspects of atmospheric ammonia (Sutton et al., 2009b). The workshop itself took place at the Scottish Government offices, with 80 experts from 19 parties to the UNECE Convention on Long-Range Transboundary Air Pollution, including support from COST729, Defra, UK Centre for Ecology & Hydrology, Scottish Government and the NitroEurope Integrated Project (CEH, 2006).

The following sections summarize key objectives of the workshop and then reflect on the developing picture, in particular considering evidence that has emerged over the last 15 years since the Edinburgh review.

Objective 1: Review of the ammonia critical level

The first objective was to assess the extent to which the existing critical thresholds for ammonia reflect current scientific understanding (Sutton et al., 2009b, p 4):

a) To examine the case for setting new ammonia critical threshold(s) based on current evidence of direct impacts of ammonia on different receptors;

b) To discuss the extent of differential sensitivity to ammonia versus other forms of reactive N;

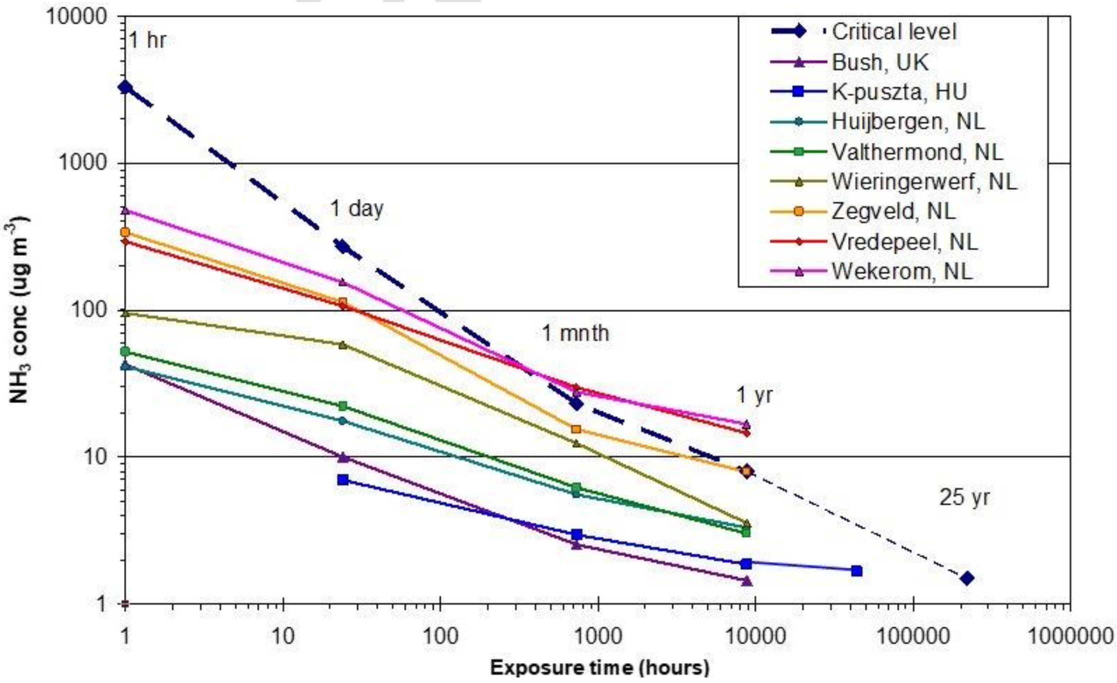
c) To debate the case for establishing indicative air concentration limits for indirect effects of ammonia, which would be consistent with current critical loads for nitrogen.

It was concluded that there was substantial evidence of long-term impacts of ammonia, such as from the Whim Bog experiment, running at that time already over 6 years, and now running for around 20 years. The most recent analyses (Levy et al., 2019; Sutton et al., 2020), emphasize that ammonia impacts integrate over periods of much longer than one year, but are also faster in occurring than the effects of equivalent amounts of wet deposited nitrogen. This provides long-term evidence that gaseous ammonia is more damaging to some forms of vegetation than wet deposited nitrogen, highlighting the importance of recognizing these impacts. It still needs to be determined if there is sufficient data available to revise critical loads based on current understanding for the differing sensitivity of dry deposited gaseous ammonia versus wet deposited nitrogen. Similarly, it was not agreed in the Edinburgh Workshop to establish indicative air concentration limits by simple equivalence to critical loads. One of the concerns here is uncertainty in dry deposition rates, especially when considering bi-directional fluxes. By contrast, adoption of the revised critical levels for ammonia has given appropriate prominence to its adverse effects.

One of the outcomes from the Edinburgh Workshop that has received least attention is the demonstrated relationship between the previous critical level and long-term ammonia monitoring data (Figure 3). This showed that the 1992 critical levels are most precautionary over longer averaging periods, compared with shorter averaging periods (Sutton et al., 2009c). Hence, in Figure 3 it can be seen that the old annual critical level is more likely to be exceeded than the hourly and daily critical levels for the NH_3 monitoring datasets examined. Again, this demonstrated the importance of setting a long-term critical level (expressed as an annual mean, but designed for application over several decades).

Figure 3: Comparison of ammonia concentration data from long term monitoring with the Egham 1992 critical levels for ammonia, as well as an extrapolated potential critical level for averaging over 25 years.

The data from multiple sites show that the critical level is most precautionary over longer averaging periods.



Source: Sutton et al., 2009c

The previous UNECE Critical Levels from the Egham Workshop 1992 were as follows:

- 8 µg m^{-3} annual mean,
- 23 µg m^{-3} monthly mean,
- 270 µg m^{-3} daily mean,
- 3300 µg m^{-3} hourly mean.

The further evidence from multiple field studies including from the UK, Switzerland, Portugal and Italy (Cape et al., 2009a,b) showed that the annual mean value was not sufficiently precautionary, and that effects accumulate for periods longer than 1 year. Data from the Netherlands were also available, but it was concluded that ambient ammonia concentrations were too high to derive a critical level from these data.

The following values were agreed in Edinburgh (Sutton et al., 2009b; Cape et al., 2009a,b) and subsequently included in the UNECE Mapping Manual (current edition: UNECE, 2017):

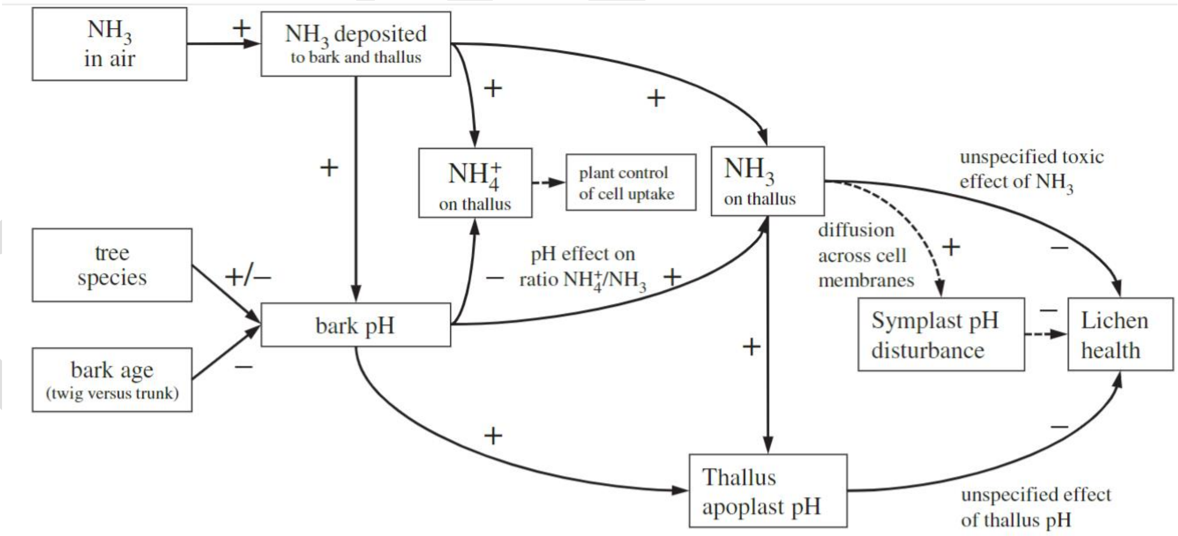
> ‘Long-term’ annual average critical level for lichens, bryophytes and for ecosystems in which they are important were set at 1 µg NH_3 m^{-3}.

> ‘Long-term annual average critical level for higher vegetation (inc. woodland ground flora): were set at 3 [2-4] µg NH_3 m^{-3}.

These critical levels were set as the long-term values, which cannot be assumed to protect for >20-30 years (expressed as annual mean). Conversely, it was concluded that there was insufficient evidence to revise the short-term critical level values (monthly, daily, hourly).

Figure 4: Possible mechanisms by which atmospheric NH_3 pollution affects epiphytic lichens, including both positive (+) and negative (−) effects.

Solid lines indicate observed relationships or those directly implied by physico-chemistry.



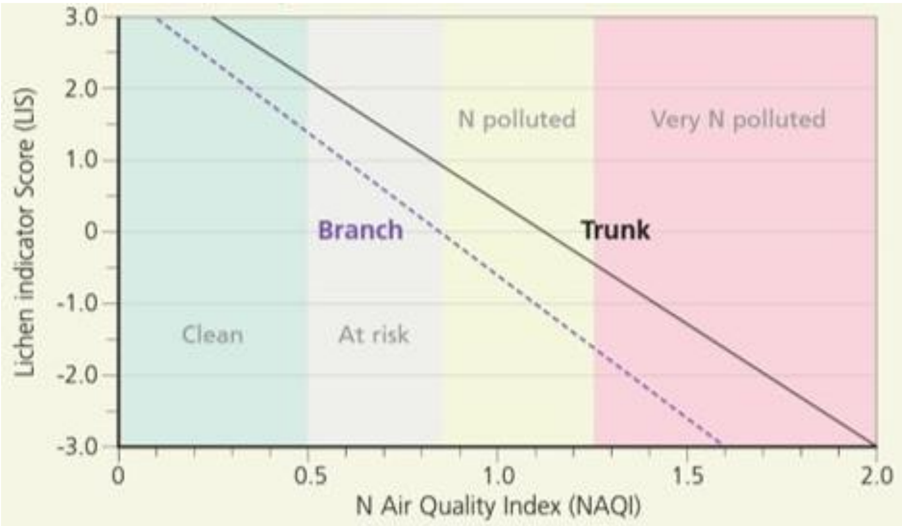
Source: Sutton et al., 2020

Subsequent analysis since 2006 has given further understanding of the potential mechanisms for why ammonia should have a more adverse effect than ammonium. This includes the observation that ammonia has an ‘alkaline air‘ effect, indicated by the observation that the pH of tree bark and other vegetation surfaces increases in the vicinity of ammonia sources (Figure 4). The relative importance of nitrogen eutrophication vs the alkaline air impacts requires further investigation.

Further evidence that needs consideration suggests that NH_3 and NO_x can have combined effects. Analysis of UK-wide data for epiphytic lichens (Lewis, 2012) identified a relationship with Lichen Indicator Score (LIS) leading to a Nitrogen Air Quality Index (NAQI), which has been applied by the UK Field Studies Council in its lichen nitrogen air quality guide (Wolseley et al. 2014). The following relationship was identified based on the UK data, and applied in Figure 5.

NAQI: Nitrogen Air Quality Index = $2[NH_3]^{0.5} + [NO_2]^{0.5}$ (using µmol m^{-3}).

Figure 5: Relationship between the Lichen Indicator Score (LIS) and the Nitrogen Air Quality Index (NAQI).



Source: Wolseley et al., 2013

Based on this NAQI relationship derived from the UK epiphytic lichen data, the following scenarios can be envisaged:

1 µg m^{-3} NH_3 and 0 µg m^{-3} NO_2	NAQI = 0.49	(Scenario 1)
0 µg m^{-3} NH_3 and 30 µg m^{-3} NO_2	NAQI = 0.8	(Scenario 2)
0 µg m^{-3} NH_3 and 11.5 µg m^{-3} NO_2	NAQI = 0.5	(Scenario 3)
1 µg m^{-3} NH_3 and 30 µg m^{-3} NO_2	NAQI = 0.49 +0.8=1.29	(Scenario 4)
0.5 µg m^{-3} NH_3 and 10 µg m^{-3} NO_2	NAQI = 0.34 + 0.47 = 0.81	(Scenario 5).

Considering the scenarios outlined above, Scenario 1 shows the value of NAQI at the ammonia critical level, which links to the change from clean to at risk in Figure 5. By comparison, Scenario 2 is the standard NO_2 critical level (NAQI=0.8), which, according to the lichen epiphyte data, is not as precautionary as the NH_3 critical level (NAQI=0.5). Scenario 3 shows that the same level of precaution as the NH_3 critical level (NAQI=0.5) would be NO_2 of around 11.5 µg m^{-3}. Scenario 4 illustrates that if both NH_3 and NO_2 are at the critical level, the overall threat to epiphytic lichens is larger (NAQI=1.29) than if only one gas is considered. Finally, Scenario 5 indicates a case where both NH_3 and NO_2 individually are less than the critical level, but the combined risk to epiphytic lichens (NAQI=0.81) is larger than having NH_3 alone at the critical level (NAQI=0.5).

These simple scenarios have implications for future consideration of how the NH_3 and NO_2 critical levels fit together. According to the UK data from which this relationship was derived, the NH_3 and

NO_2 effects are additive. Hence this suggests that a smaller critical level for NO_2 would be expected where NH_3 concentrations are high, and *vice versa*. This requires further investigation to inform future development of the critical levels approach for NH_3 and NO_2.

Objective 2: Atmospheric ammonia measurements

The second objective of the Edinburgh Workshop was to assess the extent to which independent atmospheric measurements can verify where regional changes in NH_3 emissions have and have not occurred (Sutton et al., 2009b, p 3):

a) To quantify the extent to which regional changes in ammonia emissions have been reflected in measurements of ammonia and ammonium in the atmosphere;

b) To distinguish cases where the estimated changes are due to altered sectoral activity or the implementation of abatement policies to see if atmospheric measurements verify the ammonia abatement policies;

c) To make recommendations for future air monitoring and systems for assessing the national implementation of ammonia abatement policies and consider the implications of any non-linearities.

The overall state of the evidence was summarized by Bleeker et al. (2009), showing that an 'ammonia gap' in the Netherlands - between measured and modelled NH_3 concentrations - was partly explained. Trends over time were consistent, while differences in absolute values could be attributed to typical model-measurement differences. The differences between modelled versus measured trends could also be understood by considering the case of Hungary where ammonia concentrations had not decreased despite reductions in fertilizer use and animal numbers. Comparison with aerosol and wet deposition data and a modelling scenario showed that this could be understood because SO_2 emissions had declined over the period, leading to a longer atmospheric residence time for gaseous NH_3 (Horvath et al., 2009). The implication is that ammonia monitoring needs to consider all the phases, including gaseous, aerosol and wet deposition. Subsequent reports of 20-year time series of ammonia monitoring compared with models (e.g. van Zanten et al., 2017; Wichink Kruit, 2017; Tang et al., 2018) have further improved our understanding of the relationships between ambient ammonia concentrations and changing air chemistry. In parallel, Europe-wide measurements have shown the importance of ammonia and ammonium in the overall inorganic pollution load (Tang et al., 2020).

Objective 3: Ammonia Hotspots

The third objective of the Edinburgh Workshop was to review approaches for downscaling transboundary assessments to deal with ammonia 'hotspots' (locations in landscapes with very high ammonia levels) in relation to operational modelling and monitoring (Sutton et al., 2009b, p 5):

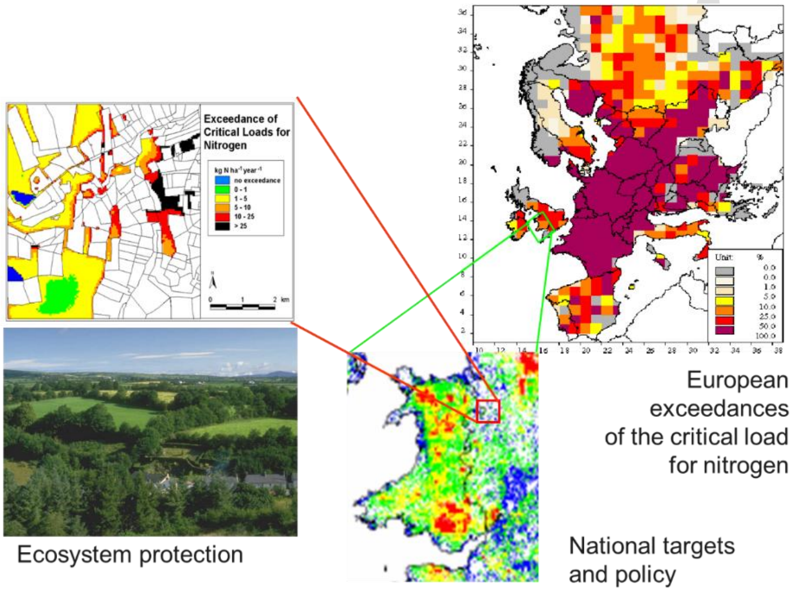
a) To review current emission and atmospheric dispersion modelling methods for downscaling NH_3 dispersion and deposition in hotspots;

b) To examine the status of methods for effect assessment and air monitoring in NH_3 hotspots;

c) To recommend broad principles for assessment of ammonia hotspots, including interactions between transboundary ammonia emission reduction targets and other policy measures.

A major review for the workshop of the challenges of assessing nitrogen in hotspots was provided by Loubet et al. (2009). Assessing ammonia in hotspots remains an issue of high uncertainty, especially emissions and dry deposition. The workshop concluded that there is a need for better knowledge on ammonia compensation points, surface resistances, and dependence on climatic variables and deposition history. It was noted that there is potential for local “landscape models” to develop local “tailored” abatement measures, as well as to quantify sub-grid variability (see Figure 6). Detailed landscape case studies have been provided by Dragosits et al. (2006), Cellier et al. (2011) and Vogt et al. (2013). Subsequent work has continued to highlight the fine scale spatial variability in ammonia concentrations, and the consequent risk for protected areas such as the Natura 2000 network (Hallsworth et al., 2010), as well as landscape-based mitigation opportunities (such as tree-belts and atmospheric buffer zones). In this case, enhanced rates of NH_3 deposition to woodland vegetation are used in landscape planning measures to increase deposition to targeted sites, e.g. non-conservation woodland, in order to help protect priority conservation sites (Bealey et al., 2016; Dalgaard et al., 2022).

Figure 6: Illustration of the challenges of dealing with spatial scale in nitrogen deposition and critical loads exceedances. The landscape case study is taken from.



Source: Dragosits et al. (2007)

Objective 4: Mesoscale modelling of ammonia

This final objective of the Edinburgh Workshop focused on reviewing mesoscale atmospheric transport and chemistry models in relation to their formulation and results for ammonia (Sutton et al., 2009c, p7):

a) To review emission parameterizations used in the models, establishing comparability, spatial and temporal resolution and uncertainties;

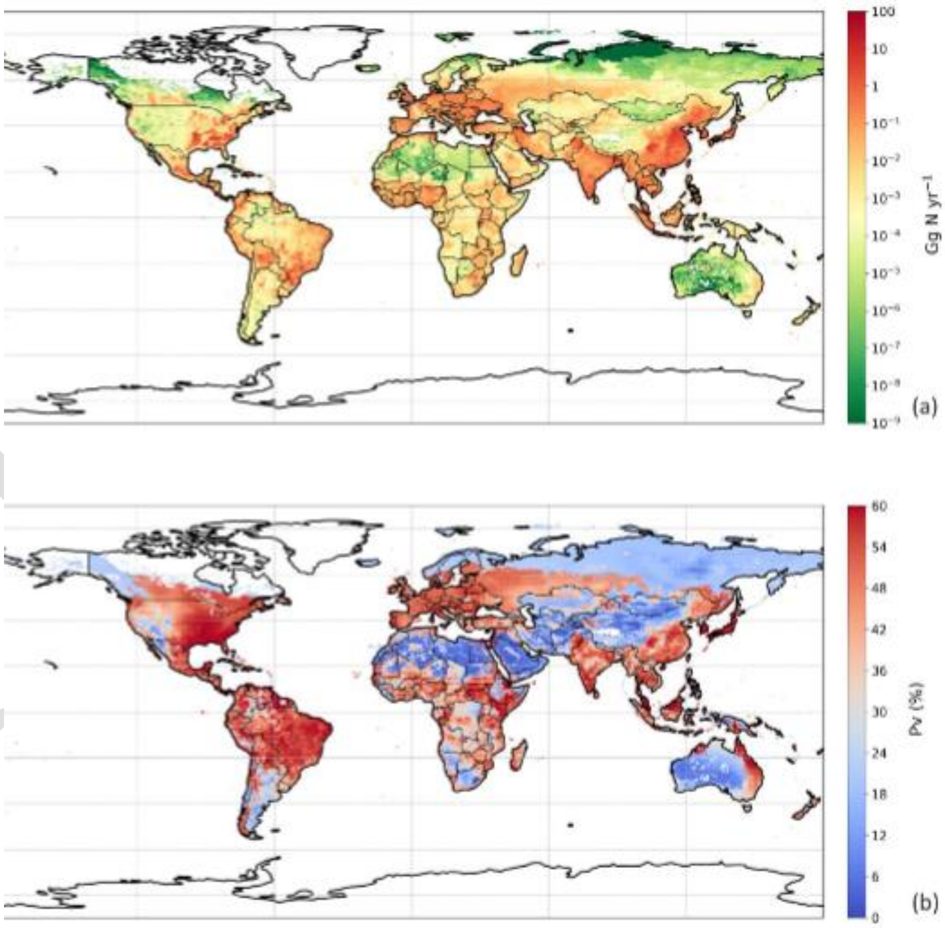
b) To review dispersion, air chemistry and deposition formulations identifying key differences and uncertainties;

c) To assess the overall performance of the models against measurements, giving model recommendations and implications of non-linearities for integrated assessment models.

A detailed review was provided for the workshop by van Pul et al. (2009). Six models from national to regional (European) scale were reviewed. This showed that estimates of wet deposition and aerosol NH_4^+ concentrations are generally adequate. It was concluded that all European scale models currently underestimate ground level NH_3 concentrations, but better agreement is found in national models. Key uncertainties were identified linked to emissions, dry deposition, spatial resolution and description of vertical diffusion. It was noted that none of the models routinely included compensation points, and a more comprehensive database was needed. This has since partly been provided by the review of Massad et al. (2010), though global scale extension is needed as evidence was predominantly from European and North American contexts. In addition to the emergence of satellite data as a key tool for monitoring ammonia trends (e.g. van Damme et al., 2018, 2021), perhaps the most significant message to be emphasized since this time is the strong climatic dependence of ammonia emissions. To date ammonia emissions inventories almost universally assume that emissions are not dependent on climate, so that emission factors are not affected by climate change. However, as illustrated by Sutton et al. (2013), Riddick et al. (2018) and Jiang et al. (2021) there is a substantial climate dependence where a warmer world is projected to emit more ammonia.

The case is clearly illustrated for poultry which excrete uric acid, where both temperature and water availability affect hydrolysis to form ammonia, and these same factors, together with windspeed also affect volatilization rates. The combined impact of these factors is shown in Figure 7 (Jiang et al. 2006). According to this mechanism NH_3 emissions could easily increase by around 30-40% over the 21st century solely due to climatic warming (Sutton et al., 2013; Riddick et al., 2018). This is of major importance in emphasizing the necessity of emission controls to avoid increasing ammonia levels and worsening the impacts on ecosystems (and human health) in future.

Figure 7: Simulated a) annual global NH_3 emissions (Gg N yr^{-1}) from poultry farming in 2010. b) Percentage of excreted nitrogen that volatilizes (PV, %) as NH_3 from chicken housing in 2010. The resolution is 0.5°´0.5°.



Source: Jiang et al. (2006)

Outlook

The Edinburgh Workshop provided a strong foundation for revising the ammonia critical level, which in the case of lichens, bryophytes and ecosystems where these are important to ecosystem integrity, reduced the critical level by a factor of eight (from 8 to 1 µg m^{-3}). Other evidence presented at the workshop has helped inform better understanding of ammonia monitoring, hotspot analysis and modelling at regional scales.

Since the Edinburgh Workshop, ongoing long-term experimentation has continued to demonstrate the sensitivity of vegetation to ammonia. Regional lichen surveying across the UK has also indicated the potential for an additive effect of NH_3 and NO_2 on lichens, which requires further investigation in the context of reviewing the NO_2 critical level.

There is an obvious need for further experimentation on the impacts of gaseous ammonia. In this context, the authors are grateful to UKRI for funding the GCRF South Asian Nitrogen Hub, which has recently established two new ammonia enrichment facilities in woodland ecosystems, one in Scotland and the other in Sri Lanka. These sites are expected to deliver improved understanding on ammonia critical levels in the future, including their effect on epiphytic lichens and bryophytes. Preliminary data from the Sri Lanka field site hosted by the Dilmah Conservation Center for Climate Change Research & Adaptation (DCC) (Queensbury Tea Estate), show that the site is highly suitable for the long-term monitoring experiment. Results showed the ammonia concentrations range from 0.4 µg m^{-3} NH_3 in the natural forest (Rilagala forest) to 1.2 µg m^{-3} at the edge of the forest (meteorological station at DCC) and 2.1 µg m^{-3} in surrounding tea plantations, as compared with 2.4 µg m^{-3} at a potato farm (Seetha Eliya). The datasets that will emerge from such new experiments are especially relevant in regions such as South Asia (Ellis et al., 2021), where limited evidence has until now been available compared with other regions.

Acknowledgements

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4.1.2 Literature review on the effects of ammonia

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This presentation summarized the main findings of a background document that was prepared before the workshop and was sent to the participants. It focussed on early and more recent ammonia research and concluded that during recent years not many experiments with regard to the effect of ammonia on plants had been conducted compared to earlier years. The update of the review is presented in chapter 2 of the present document and includes several additions and modifications that were communicated to us after the workshop.

4.1.3 Modelling ammonia concentrations, trends and scenarios in Europe

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Whilst NH_3 emissions in Europe has been reduced only by ca 12% during the 2000-2019 period, much larger emission reductions have been reported for SO_x (ca 80%) and NO_x (ca 50%) emissions. Using the regional scale EMEP/MSC-W model, we have analyzed how these emission changes affected concentrations and wet depositions of reduced nitrogen during this period (Aas et al. 2021). Furthermore, we have studied the effects of reductions in NH_3 emissions on $PM_{2.5}$ concentrations and depositions of nitrogen in Europe in the light of present (2017), past (2005), and future (2030) conditions (Jonson et al. 2022).

Few observational sites show statistically significant trends in wet deposition of reduced nitrogen in model results or EMEP observations for the 2000-2019 period. On the other hand, total ammonium in air (NH_3+NH_4^+) decreased by about 25% in observations and model calculations for the same period, which is larger than the reduction in ammonia emissions. The reduction for ammonium aerosols is even larger, around 50%, both in observations and model calculations. These large differences between the changes found for the different reduced nitrogen components can be explained by the interaction of ammonia with the sulfur and oxidized nitrogen components. Furthermore, the results imply that the contribution of ammonia emissions to aerosols has been largely reduced during the 2000-2019 period, due to the impact of SO_x and NO_x emission reductions.

Assuming that the NEC Directive will be met by 2030, we calculate that reductions in $PM_{2.5}$ levels per gram of NH_3 emissions mitigated are about a factor of 2.6 lower starting from the level of 2030 emissions compared to using 2005 emissions (due to the less efficient formation of NH_4^+ when SO_x and NO_x emissions have been reduced much more than NH_3 emissions). Thus, mitigation of NH_3 emissions are expected to reduce $PM_{2.5}$ less efficiently than it used to.

Following the expected reductions of NH_3 emission (19 % reductions in 2030 compared to 2005), depositions of reduced nitrogen will also decrease in Europe. However, as the reductions in NO_x emission are larger than for NH_3, the fraction of total nitrogen (reduced plus oxidized nitrogen) deposited as reduced nitrogen is increasing, and may exceed 60% in most of Europe by 2030. Thus, the potential for future reductions in the exceedances of critical loads for eutrophication in Europe will mainly rely on the ability to reduce NH_3 emissions.

A full description of these studies can be found in Aas et al. (2021) and Jonson et al. (2022).

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4.1.4 Measuring ammonia in the Netherlands

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Why do we monitor?

The National Institute of Public Health and Environment (RIVM) in the Netherlands is responsible for different environmental monitoring networks. For air quality the two relevant networks for ammonia measurements are the National Monitoring Network Air Quality (LML) and the network Monitoring Ammonia in Nature Areas (MAN).

The overall measurement objectives for the ammonia measurements are:

- Model validation and calibration
- Determining trends in time
- Performing process studies
- Reference function for other related measurements

In the next sections, a short overview is given of the different ammonia measurements in the Netherlands.

Intensive monitoring of ammonia concentrations

There are six high-accuracy ammonia measurement sites within the LML network (Figure 8). By means of miniDOAS systems (Berkhout et al., 2017), where DOAS stands for Dual Optical Absorption Spectroscopy, hourly concentrations of ammonia are collected. The miniDOAS is an open path optical system, with no tubing involved. This has the advantage that there is no delay due to NH_3 sticking to inlet lines or air filters. Furthermore, such an optical system has no interference with ammonia salts. Information on these measurements can be found at https://www.rivm.nl/landelijk-meetnet-luchtkwaliteit (in Dutch).

Low-cost monitoring of ammonia concentrations

The low-cost monitoring of ammonia concentrations in the Natura2000 network in the Netherlands consists of more than 500 samplers at more than 300 locations distributed over the country (Lolkema et al., 2015; Noordijk et al., 2020; Figure 8). These locations are within 86 Natura 2000 areas. The measurements are performed by means of Gradko passive samplers. Monthly concentrations are collected for about 2-9 measurement locations per Natura 2000 area, with at least one triplicate per area.

The overall advantages of the low-cost passive samplers are:

- They are small
- They are cheap
- No housing and/or electricity needed
- Monthly sampler change is easy (so samplers can be replaced by local assistants, enabling a large spatial distribution)

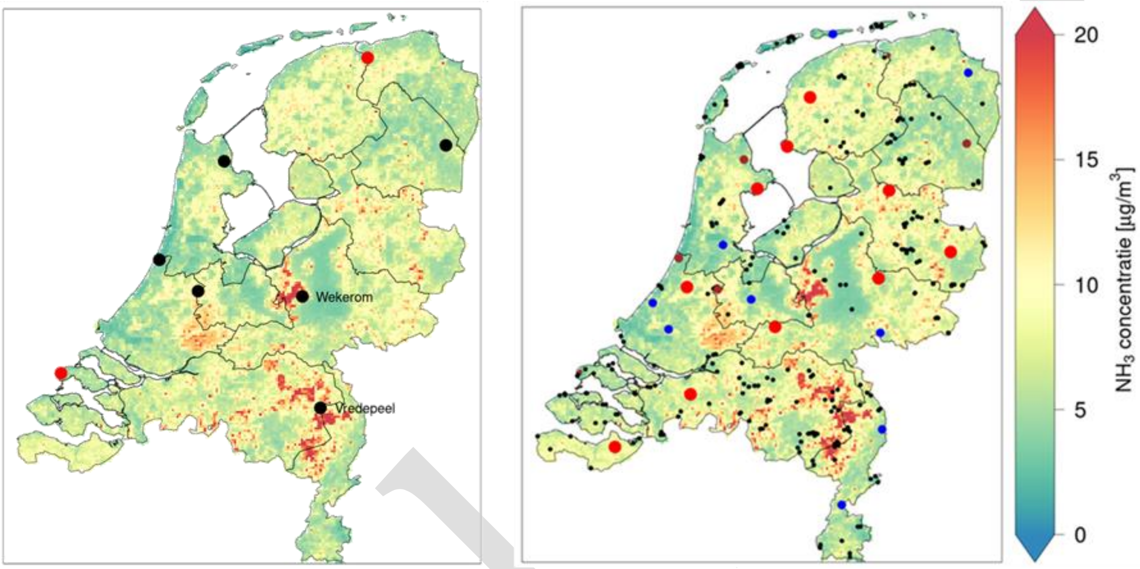
However, there is also an important disadvantage: the measurements are less accurate than high-accuracy instruments.

To largely overcome this accuracy issue, a hybrid design is used in the Netherlands. In this design, low-cost samplers and high-accuracy data from the LML stations are combined allowing a calibration correction on a monthly basis.

Information on the low-cost measurements in nature areas, including the monitoring results per Natura 2000 area, is available via https://man.rivm.nl (in Dutch).

Figure 8: Spatial distribution of intensive high-resolution miniDOAS (left) and low-cost Gradko (right) measurements in the Netherlands.

Black are current measurement locations, in red and blue the proposed locations for expansions.



Source: Wichink Kruit et al. (2021).

Dry deposition monitoring of ammonia

In the Netherlands, a small network exists for monitoring of dry deposition of ammonia. By means of a gradient technique, the dry deposition is determined by combining the concentration different between two heights and a measure of turbulence. At four locations in the Netherlands these measurements are performed using a COTAG system (Conditional Time Averaged Gradient, e.g. Famulari et al., 2010), providing monthly depositions. Information on these measurements, including the monitoring results, can be found at www.rivm.nl/stikstof/meten/drogedepositieNH3 (in Dutch).

Research is ongoing with respect to another gradient system. Two miniDOAS systems are used in a gradient setting, providing half-hourly dry deposition values (Swart et al., in preparation).

Development in the Dutch networks

In the coming year(s) different expansions of the measurements are scheduled in the Netherlands (Wichink Kruit et al., 2021; in Dutch):

- The number of intensive miniDOAS systems will be increased from six to eight, with the two additional stations located in low concentration areas near the Dutch coast (Figure 8).

- The number of low-cost Gradko measurements will be expanded by a total of 19 locations; with ten agricultural locations and nine urban locations and at country borders (Figure 8).
- The COTAG dry deposition network will be expanded from four to ten locations, spatially distrusted over the Netherlands and covering different ecosystem types.
- Furthermore, the measurements of ammonium and nitrate aerosols in the air and wet deposition of reduced and oxidized nitrogen will be expanded.
- Not only the 'traditional' monitoring activities are expanded, also the use of satellite data will be explored.

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Wichink Kruit, R. et al., 2021. Op weg naar een optimale meetstrategie voor stikstof, Rijksinstituut voor Volksgezondheid en Milieu, Bilthoven.

4.1.5 Continuous monitoring of ammonia concentrations with Cavity Ring-Down Spectroscopy

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High-precision trace gas measurements are essential for a range of industrial and air quality monitoring purposes. Picarro's Cavity Ring-Down analyzers (CRDS) have become the reference technology for continuous, high precision trace gas measurements, and they are used for numerous gas monitoring purposes like measuring continuously the ammonia concentration in ambient air, including *in situ* studies (Twigg et al. 2022). Key features of Picarro's CRDS analyzers are: (i) low calibration requirements, (ii) field deployable, (iii) negligible interference ('interference-free'), (iv) long term unattended operation, and (v) the possibility to measure multiple species. Here we present the performance and use cases for the Picarro G2103 NH_3 analyzer (e.g. Kamp et al. 2019) as well as the recently released Picarro G2509 multi species analyzer that allows to measure NH_3, CH_4, CO_2, N_2O and H_2O simultaneously. For NH_3, these analyzers reach a precision in the ppt range.

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4.1.6 Ammonia monitoring at long-term and temporary sites in Flanders

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Flanders Environment Agency (VMM) has been monitoring ambient ammonia (NH_3) since 2002. The network currently consists of 17 sites located in nature areas and rural areas closer to known NH_3 sources. The air concentrations are determined using diffusive samplers and reported according to EN 17346 (2020). The main aims of the measurements are to assess annual mean NH_3 levels, to examine time trends and to calibrate and validate results of atmospheric dispersion modelling.

In 2020, the annual mean NH_3 was lower than the critical level for higher plants (3 µg/m^3) at only 4 of 17 sites. The critical level for lichens and bryophytes (1 µg/m^3) was not reached. Since 2008, on the average no decrease in the measured annual site means has been observed. To estimate NH_3 at the regional level, the Flemish version of the Operational Priority Substances model is applied (Sauter et al. 2018) at a grid cell resolution of 1 km^2. The modelled annual NH_3 is calibrated based on the measurements using a single linear regression approach.

Flanders Environment Agency also carries out temporary measurement campaigns. In 2015-2016, NH_3 was measured at 106 extra sites in nature areas. This study indicated that the permanent monitoring sites were spatially representative for the background NH_3 concentrations in Natura 2000 sites in Flanders (VMM 2017). The model slightly overestimated the measurements, but there was a good linear correspondence between the model and measurements results. In 2017, NH_3 was measured at 60 temporary sites in 6 nature areas to examine local variations. This study showed that the modelling results could be improved by combining the regional OPS model outcomes with local modelling near sources by the Immission Frequency Dispersion Model (IFDM).

Because of the important contribution of NH_3 to nitrogen deposition and secondary particulate matter, there has been increasing interest in this pollutant. In this respect, VMM investigated the possibilities of more innovative techniques such as differential optical absorption spectroscopy (DOAS) and the COnditional Time Averaged Gradient (COTAG) method. The DOAS method is an open-path technique that allows real-time measurements of NH_3 at high temporal resolution without sampling artefacts. In collaboration with RIVM we have been using two miniDOAS monitors since 2017. Recently we also set up two COTAG devices at heathland sites to determine the net dry deposition flux of NH_3, in collaboration with UKCEH.

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4.1.7 Monitoring atmospheric ammonia on Natura2000 sites in the Republic of Ireland

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The Republic of Ireland has exceeded its ammonia (NH_3) emission ceiling under the National Emission Ceilings (NEC) Directive (2016/2284/EU) (European Union 2016) since 2016 (Environmental Protection Agency 2021). Additionally, Ireland's national ambient ammonia monitoring carried out in 2013 – 2014 across 26 sites only identified three below the critical level of 1 µg NH_3 m^{-3} (Doyle et al. 2017). It was hence deemed appropriate to carry out targeted atmospheric ammonia monitoring on Natura 2000 sites protected under the Habitats Directive (92/43/EEC) (European Economic Committee 1992). From August 2017 through to July 2018, atmospheric ammonia was monitored across 12 Natura 2000 sites in the Republic of Ireland using ALPHA samplers in triplicate (Kelleghan et al. 2021a). A subset of Natura 2000 sites containing potentially sensitive qualifying interests were prioritised. Table 3 lists habitats included alongside the range of monitored atmospheric concentrations and modelled total nitrogen deposition rates for each habitat type. This work monitored airborne concentrations of 0.47 – 4.59 µg NH_3 m^{-3} across all sites. These monitored concentrations were used to calculate the dry deposition of NH_3 based on habitat specific deposition velocities (de Kluizenaar and Farrell 2000). Other forms of deposited nitrogen were extracted from EMEP modelling (The Norwegian Meteorological institute 2019) and summed to estimate total nitrogen deposited.

Table 3: Summary of atmospheric ammonia (NH_3) monitoring and modelled nitrogen (N) deposition across 12 Natura 2000 sites during 2017-2018 in the Republic of Ireland

No. of sites	Qualifiying Feature	NH_3 concentration / µg NH_3 m^{-3}	N Deposition / Kg ha $year^{-1}$
6	Active raised bogs*	1.18 – 2.92	10.1 – 17.32
1	Blanket bogs (active)	0.57	9.52
1	Transition mires and quaking bogs	1.40	11.07
2	Semi-natural dry grasslands and scrubland facies on calcareous substrates (*Festuco-Brometalia*) (* important orchid sites)	0.47 – 2.29	5.93 - 12
2	Old sessile oak woods with *Ilex* and *Blechnum* in the British Isles	2.7 – 3.1	13.64 - 15.27
1	Hard oligo-mesotrophic waters with benthic vegetation of *Chara* spp.	4.59	17.78

Considering Active raised bogs* are a priority Annex I habitat within the Habitats Directive (92/43/EEC) and the sensitivity of *Sphagnum* moss to atmospheric ammonia, the recorded exceedance of both their critical level (1 µg NH_3 m^{-3}) and empirical critical load (5 – 10 Kg ha $year^{-1}$) is cause for concern. Recent national modelling has also shown that modelled NH_3 concentrations across all active raised bog SACs in Ireland exceed their critical level (Bealey et al. in press). Where the Blanket bogs (active) site does not exceed its critical level for NH_3, when other forms of nitrogen are added, it exceeds the lower range of its empirical critical load (5 – 10 Kg ha $year^{-1}$), highlighting the need to consider both critical levels and loads during assessment of impacts. While one Semi-natural dry grasslands and scrubland facies on calcareous substrates (*Festuco-Brometalia*) (* important orchid sites site fell below both its critical level and load, the other exceeded its critical level and its vegetation community change point (8.3 Kg ha year^{-1}) (Wilkins et al. 2016). Both relevant critical levels and loads were exceeded in all remaining SACs. Monitoring identified strong statistical relationships between monitored NH_3 and both the proportion of agricultural land within 2 km (Pearson correlation co-efficient of 0.8) and temperature (Pearson correlation co-efficient of 0.9). The relationship between temperature and ammonia concentration is potentially confounded by seasonally variable agricultural practices. This monitoring also showed a positive correlation with the MARSH (Mapping Ammonia Risk on Sensitive Habitats) model (Kelleghan et al. 2019) with a Pearson correlation co-efficient of 0.7 (Kelleghan et al. 2020). Kelleghan et al. (2020) clarified that, on at least one site (Raheenmore Bog SAC with an annual average concentration of 2.3 µg NH_3 m^{-3}) ecological indicators of atmospheric NH_3 pollution were observed. Such indicators are displayed in Figure 9 including, proliferation of nitrophytic species (*Xanthoria parietina* and algae) and breakdown and decay of both *Sphagnum* sp. and *Cladonia portentosa*. This site was in receipt of NH_3 primarily from local cattle production and fertiliser/slurry application (Kelleghan et al. 2020).

Figure 9 Ecological indicators of ammonia pollution on peatland site in the Republic of Ireland

Source: Fotos from David B. Kelleghan

The National Ecosystem Monitoring Network (NEMN) is being developed in the Republic of Ireland as a response to the updated NEC Directive (Kelleghan et al. 2021b), which requires monitoring of air quality impacts across sensitive habitats. Such networks are required by the European Commission to be representative, risk-based and cost-effective. Linking with and utilising monitoring carried out by other networks is key to the success of the NEMN. Additionally, adoption of a tiered monitoring design adapted from ICP Forests monitoring minimises the cost of such a network while maximising its benefits. This design requires numerous Level 1 sites (15 minimum per habitat) where only non-invasive periodic monitoring occurs. This monitoring is dependent on other networks, such as that carried out under Article 17 of the Habitats Directive (92/43/EEC) every six years or ICP Forests crown condition surveys every four years.

The addition of parameters such as soil sampling and percent nitrogen tissue analysis (of moss species) are currently being explored, alongside inclusion of permanent quadrats (within Article 17 monitoring). Level 2 sites require some level of continuous passive atmospheric monitoring. NEMN proposes splitting these into Level 2 and Level 2 core, where Level 2 sites focus primarily on NH_3

monitoring and wet deposition where required and Level 2 core sites are intended to receive a full suite of atmospheric monitoring. This would include use of passive samplers for NH_3 and NO_X, bulk precipitation and active denuders for gases and aerosols; additional monitoring can be added as the network develops. The current NEMN design aims to have at least two Level 2 core sites per habitat, expanded by Level 2 sites to increase coverage across the risk gradient. As this is a tiered design, it is intended that each Level 2 core site will also receive Level 2 monitoring, and each Level 2 site will also function as a Level 1 site. This tiered approach ensures that relationships between atmospheric ammonia and nitrogen deposition with ecological responses can be quantified. This is an iterative network, intended to grow from year to year to monitor impacts long-term impacts. Data collected will be key to assessing and developing habitat-specific critical levels and loads.

Researchers in the University College Dublin School of Biosystems and Food Engineering have been communicating the effects of atmospheric NH_3 to the general public in Ireland through television, radio and newspaper. Their most recent endeavour is a comic suitable for children about an anthropomorphic cloud of ammonia gas. "There's Something about Ammonia" (Wright et al. 2021) gives a basic introduction to sources, impacts and solutions to NH_3. This work has thus far been translated to 15 languages thanks to the assistance of the EPA, the COST-Action LivAge and a number of volunteers. This comic is freely available to download and use (https://www.ucd.ie/ammonian2k/publications/comic/).

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4.1.8 National ammonia monitoring network in the UK: its current use and future potential

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The Department for Environment, Farming and Rural Affairs (Defra) supports work to monitor concentrations of gaseous ammonia (NH_3) and particulate ammonium (NH_4^+) in the UK. The UK Centre for Ecology and Hydrology (UKCEH) manages the National Ammonia Monitoring Network (NAMN) for Defra.

The NAMN was established in 1996 with the objective of quantifying temporal and spatial changes in air concentrations and deposition of NH_3 and NH_4^+ (included since 1999) on a long-term basis[4]. The monitoring provides a baseline to verify compliance with international agreements, such as the UNECE Gothenburg Protocol 1999 and the EU National Emissions Ceilings Directive NECD 2016/2284 (superseded in the UK by the National Emissions Ceilings Regulations 2018 since EU exit).

The National Ammonia Monitoring Network (NAMN) is part of the UK Air Pollution Impacts on Ecosystem Networks (APIENs). UK APIENs was formed in 2018 by integrating national air quality and ecosystem monitoring networks and surveys to meet monitoring (Article 9) and reporting (Article 10) obligations under the EU National Emissions Ceilings Directive (NECD 2016/2284). Since the UK exited the EU reporting has been transposed under Part 5 of the UK National Emissions Ceilings Regulations 2018 and is aligned to the 4 yearly European reporting cycle. APIENs is designed to monitor and report the negative impacts of air pollution (e.g., acidification, eutrophication, ozone damage and changes in biodiversity) on ecosystems that are representative of freshwater, natural and semi-natural habitats and forests in the UK[5].

The UK ammonia monitoring network has sites located across representative areas across the UK (Figure 10). There are 52 ALPHA© and 27 DELTA© monitoring sites (Table 4) collecting spatial and temporal changes in ammonia and ammonium concentrations and deposition on a long-term basis. It is important to note that the DELTA© samplers also collect Nitric acid (HNO_3), Sulphur dioxide (SO_2), Hydrogen chloride (HCl), Nitrate (NO_3^-), Sulphate (SO_4^{2-}), Sodium (Na^+), Calcium (Ca^{2+}), Magnesium (Mg^{2+}) as part of the UK Acid Gas and Aerosol Network (AGA-Net)[6].

The integrated data collected will provide evidence to determine the state of the ecosystem and provide a baseline against which to measure any changes and potential recovery in ecosystem responses to emissions reductions.

[4] UKCEH Ammonia Network http://www.pollutantdeposition.ceh.ac.uk/content/ammonia-network [accessed 23/05/2922]
[5] APIENs Home page http://www.apis.ac.uk/apiens [accessed 12/05/2022]
[6] Defra UK Air Information Resource: UK Acid Gas and Aerosol Network https://uk-air.defra.gov.uk/networks/network-info?view=aganet#:~:text=The%20network%20provides%20a%20long,of%20acidity%20and%20nutrient%20nitrogen. [accessed 24/05/2022] AGA-Net is managed by the Environment Agency on behalf of Defra.

Figure 10 Distribution of UK National Ammonia Monitoring Network sites

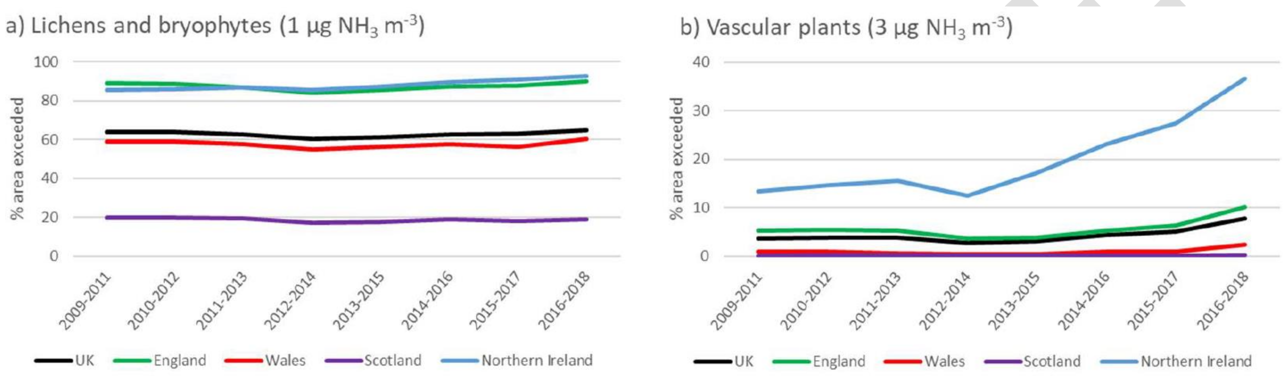
Source: CEH Edinburgh

Table 4: The distribution of ammonia network sites by UK country (source: UKCEH)

	ALPHA (NH_3)	27 DELTA (NH_3 and NH_4^+)
England	37	13
Northern Ireland	1	2
Scotland	12	9
Wales	2	3
Total	52	27

Rowe et al. (2021) reported that in 2017 for 64.7% of the UK land area ammonia concentrations exceeded the 1 µg m^{-3} critical level (set for the protection of lichens and bryophytes) compared to 63.9% in 2010 (Figure 11a). This exceedance varies spatially, with Scotland having below 20% exceedance, and England and Northern Ireland having 90% or more land area exceeding this critical level. In Northern Ireland nearly 40% of the land area exceeds the 3 µg m^{-3} critical level (set for the protection of vascular plants), rising rapidly since 2013 (Figure 11b). England and Wales have also seen increases in the proportion of their land area exceeding the 3 µg m^{-3} critical level.

Figure 11: Percentage of UK land area with ammonia concentrations exceeding critical levels: for a) lichens and bryophytes, b) vascular plants.



Source: Rowe et al. (2021)

These trends in ammonia emissions are reflected in the air pollution inventories for each of the UK countries (National Atmospheric Emissions Inventory, 2021). The normalised trends show that ammonia is the only pollutant that is significantly higher than at the 2005 reference date, especially for Northern Ireland, England, and Wales.

The UK has reported not meeting the 2020 National Emissions Ceilings Regulations ammonia target (Defra, 2022). This then triggers a review of the UK National Air Pollution Control Programme (NAPCP) to meet the 2030 target.

In recent years there has been extensive ammonia monitoring work in Northern Ireland. In addition to the three main NAMN sites, 25 ALPHA© and 4 DELTA© sites were established by DAERA in collaboration with the Agri-Food and Biosciences Institute (AFBI) and UKCEH. The objective of this project is to validate modelled NH_3 concentrations. These sites are located to achieve representative coverage geographically across locations with both high and low ammonia background concentrations. Work is underway to incorporate these data into the NAMN in the future. Some of these sites are located close to or within designated conservation sites such as Special Areas of Conservation (SAC) or Areas of Special Scientific Interest (ASSI) (AFBI, 2021).

In addition to the NI-wide network, there is an ongoing focused regime of ammonia monitoring by DAERA NIEA in collaboration with UKCEH and Ulster Wildlife (UW) on eight Special Areas of Conservation, with between 1 and 9 ALPHA© monitors on each site. Wet deposition bulk-rain collectors, NOx monitoring and vegetation analysis for foliar N levels has also been initiated on selected sites (Tang et al. 2022).

In addition to monitoring, nitrogen profiles have been developed to consider the nitrogen threats across the NI designated site network and to inform conservation management. It is hoped that this suite of work can be used to improve conservation site management through a better understanding of the ammonia and nitrogen inputs at such sites.

Pending funding, Wales plans to increase ammonia monitoring with a further 24 ALPHA© and three DELTA© sites. These will be in conjunction with local monitoring campaigns and site habitat surveys. These data will be used to confirm the background concentrations of ammonia and deposition of nutrient nitrogen. It is planned to develop nitrogen and ammonia profiles and hence sensitivity for designated conservation areas to improve conservation management. There are other examples in England where Shared Nitrogen Action Plans (SNAPs) with ammonia and nitrogen data are being used to develop conservation action plans (Natural England 2018).

The existing NAMN through the establishment of APIENs is used to provide data for the UK to report under Part 5 of the NECR. With APIENs it is intended that the integrated data can be used to monitor ecosystem recovery and input into air pollution recovery strategies. Examples have described briefly how the data from the NAMN can be used to develop strategies for controlling ammonia and help conservation management.

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4.1.9 Ammonia concentrations measurements in Germany – an overview

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National legal and policy situation - Current status in Germany

One of the most important regulations for air quality in Germany is the EU Air Quality Directive (AAQD) and its national implementation under the 39. Federal Immission Control Act (39. BImSchV). It includes requirements for the protection of vegetation elements and crops by setting target or critical values and long-term objectives for ozone, NO_x and SO_2. Measurements have to be carried out in the rural and rural background station categories. Despite known adverse effects of ammonia (NH_3) on the vegetation and ecosystems, there is no NH_3 regulation neither in the AAQD nor in the German federal legislation, yet, to control NH_3 concentrations. In contrast, the EU Directive on the reduction of national emissions of certain atmospheric pollutants (NEC Directive) and its national implementation, the 43. Federal Immission Control Act (43. BImSchV), set nationally binding emission reduction commitments for NH_3 in order to support the reduction of ambient NH_3 to the Critical Levels of the Geneva Air Quality Convention (Recital 8 of the NEC Directive). However, although effect monitoring (of e.g. eutrophication, acidification, nutrient balances, nitrogen deposition and ozone damage) is an essential part of the Directive and its national implementation, neither the NEC Directive itself, nor the national monitoring concept contain a regulation or a guidance on the monitoring of NH_3 concentrations.

In Germany NH_3 concentration regulations are only part of the project licensing processes. Within the air quality control policies for a license of a planned project (e.g. livestock housing), the planning institution has to proof that an additional project specific NH_3 load of 2 µg m^{-3} must not be exceeded. If a Natura2000 license is required, it has to be proofed that the new project, together with the background concentration, does not lead to an exceedance of the Critical Levels.

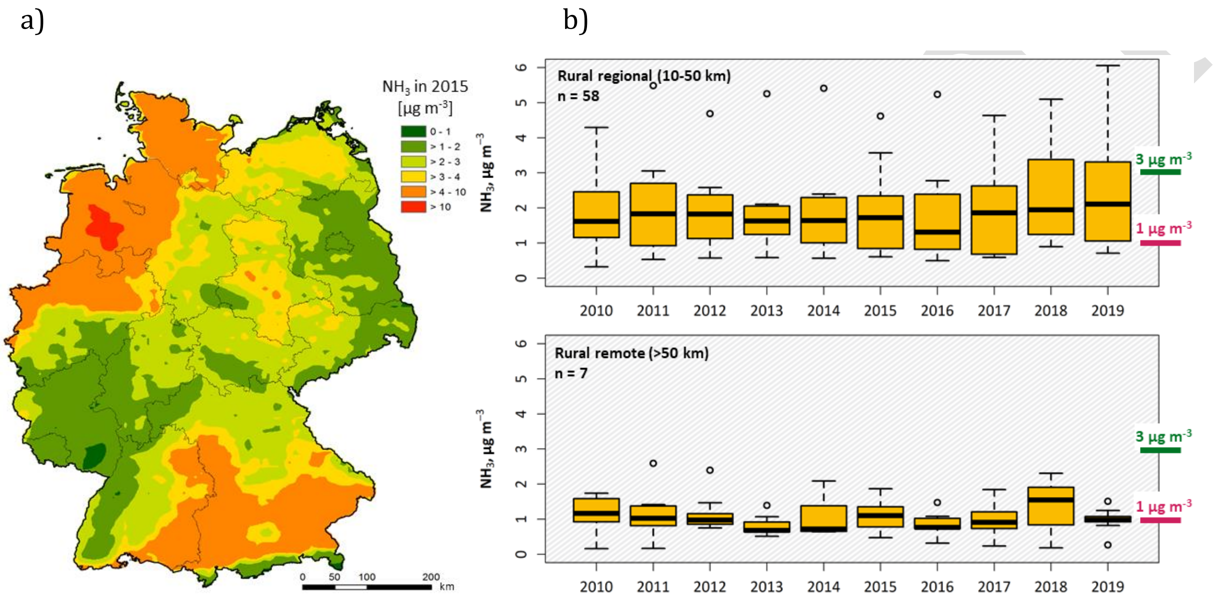
NH_3 concentrations in Germany – Status Quo

There is evidence that the CLTRAP Critical Levels for NH_3 are exceeded over large areas in Germany. Figure 12a shows the modelled surface NH_3 concentrations for an exemplary year. It is evident that in the hotspot regions of North-West and South Germany the Critical Level of 3 µg m^{-3} for higher plants is exceeded extensively on an annual basis. For the rest of Germany, the model indicates that the 1 µg m^{-3} Critical Level for lichens and moss is exceeded as well for most regions, with only a few areas where the annual NH_3 concentration is below that level. Comparison of the modelled NH_3 distribution with satellite observations (not shown) reveal that the model identifies the NH_3 hotspot regions well, however, a systematic underprediction in some areas of Eastern Germany is apparent. In-situ measurement of NH_3 are therefore crucial to evaluated and improve atmospheric models. At the same time, more extensive *in-situ* measurements are important for local pollution assessments.

In Germany, a national NH_3 monitoring network that focuses on ecosystem protection does not exist. However, numerous NH_3 monitoring sites were established by the federal states, pre-dominantly within the last 15 years, although no requirement to report NH_3 concentrations exists.

Measurements are performed by different institutions and are sometimes collected within other networks. To our knowledge, NH_3 measurements are currently performed by 11 out of 16 states at more than 93 measurements sites, predominantly in rural or remote locations. In addition, the German Environment Agency measures NH_3 at seven remote background sites across Germany. At most sites, NH_3 measurements are performed using passive samplers with a biweekly or monthly sampling interval, the network of the German Environment Agency employs denuders with a weekly sampling interval. The state of Hesse operates five sites with online measurements of NH_3 using chemiluminescence analyzers at a 30 min resolution.

Figure 12: a) Map of annual mean NH_3 surface concentrations over Germany (LOTOS-EUROS simulation for 2015; source: PINETI-3 project of the German Env. Agency). b) Annual box plot statistics of NH_3 *in-situ* measurements from 2010-2019 for regional (upper panel) and remote (lower panel) locations.



Source: Own illustration. Map (a) is based on data from the PINETI-3 project of the German Environment Agency. Data for trends (b) are taken from the air quality database of the German Environment Agency (www.env-it.de/stationen/public/open.do), which includes about 2/3 of NH3 measurement sites in Germany.

Evaluation of the *in-situ* measurements confirms the evidence from the model (Figure 12b) that the Critical Level of 1 µg m^{-3} is exceeded regularly at most sites, even at those in remote locations. The Critical Level of 3 µg m^{-3} is exceeded occasionally, strongly depending on the region. Measurements over the last decade reveal that NH_3 concentrations have been overall slightly increasing as it has been reported for other countries in Europe as well. One reason is the relative increase of NH_3 emissions compared to those of nitrogen oxides (NO_x) and sulfur dioxide (SO_2). Since the latter two act as acids in the atmosphere, their relative depletion shifts the gas to-particulate phase equilibrium to gas phase NH_3.

Relationship between Critical Loads into Critical Levels

For the monitoring of indirect effects of NH_3, such as contribution to eutrophication and related species composition change, nitrogen deposition is evaluated against ecosystem specific Critical Loads. As nitrogen deposition flux measurements (including NH_3) are challenging to make and not yet suitable for widespread monitoring, NH_3 concentration measurements could act as a proxy with a higher spatial coverage. Using a typical fraction of NH_3 dry deposition (F_{drydep_NH3-N}) to the total nitrogen deposition ($F_{totaldep\_N}$) and a typical NH_3 deposition velocity (V_{dep_NH3}) – e.g. from model evaluations – the Critical Load (CL) can be transferred to an NH_3 concentration threshold (C_{NH3_max}):

$$C_{NH3\_max} = \frac{F_{drydep\_NH3-N}}{F_{totaldep\_N}} \cdot \frac{CL}{V_{dep\_NH3}}$$

If possible, the effective deposition velocity considering NH_3 bi-directional exchange is taken. As an example, for a nitrogen sensitive ecosystem forest with $CL = 10\ kg(N)\ ha^{-1}a^{-1}$, $V_{dep\_NH3} = 1.5\ cm\ s^{-1}$ and $F_{drydep\_NH3-N}/F_{totaldep\_N} = 0.3$ yields $C_{NH3\_max} = 0.8\ \mu g\ m^{-3}$. That means that at a site with a Critical Load of 10 kg N ha^{-1} a^{-1} with the given assumptions a measured concentration of >0.8 µg $m\text{-}^{3}$ would indicate an exceedance of the Critical Load value.

The given example shows that critical NH_3 concentration values [µg m^{-3}] resulting from the conversion of ecosystem specific Critical Loads [kg ha^{-1} a^{-1}] are in the same range as the current NH_3 Critical Levels, which also has been shown initially by Cape et al. (2009). This finding gives another argument for the scientific validity of the Critical Levels in the given range. Also, the conversion offers a potential way to derive more ecosystem specific Critical Levels in comparison to current values for the aggregated groups of higher plants und the group of lichens and bryophytes.

Table 5: Considerations for an NH_3 regulation in the EU AAQD

Motivation & advantages	Difficulties & open questions
NH_3 affects ecosystems and $PM_{2.5}$ formation	NH_3 dispersion is characterised by small scale heterogeneity
A harmonization of emission reduction and air quality control could be achieved for (like for NO_x or PM) and European air quality policies could be further aligned nature protection policies	The selection of representative measurement sites is yet to be defined. Most preferably, chosen sites would represent nitrogen sensitive ecosystems. Where appropriate, in Natura2000 areas and make use of existing monitoring stations (EMEP or AAQD); affected and non-affected ecosystems
Through multiple measurements across Europe the scientific knowledge of NH_3 distribution, ecosystem effects and $PM_{2.5}$-formation would systematically be improved. This knowledge would enhance the development of effective NH_3 reduction strategies and help the setting standards and the improvements of models	It is yet to be defined, if NH_3 would be regulated with a target value, a limit value or a critical level or if a monitoring in a first phase could be made mandatory without defining a concentration level.

Current revision of the European Ambient Air Quality Directives: Window of opportunity

With regard to the current revision process of the European Ambient Air Quality Directives currently there is a window of opportunity to include a regulation on NH_3 monitoring in the future international framework legislation. Not making use of this option would lead to the unfavorable alternative, that NH_3, one of the most concerning air pollutants of our time, would remain unmentioned in the most important European regulation for ambient air quality. Some considerations have been put together in Table 5. Against the background of those consideration the German Environment Agency strongly supports the uptake of an NH_3 regulation to the revised AAQD.

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4.1.10 Metrological aspects to support environmental ammonia monitoring

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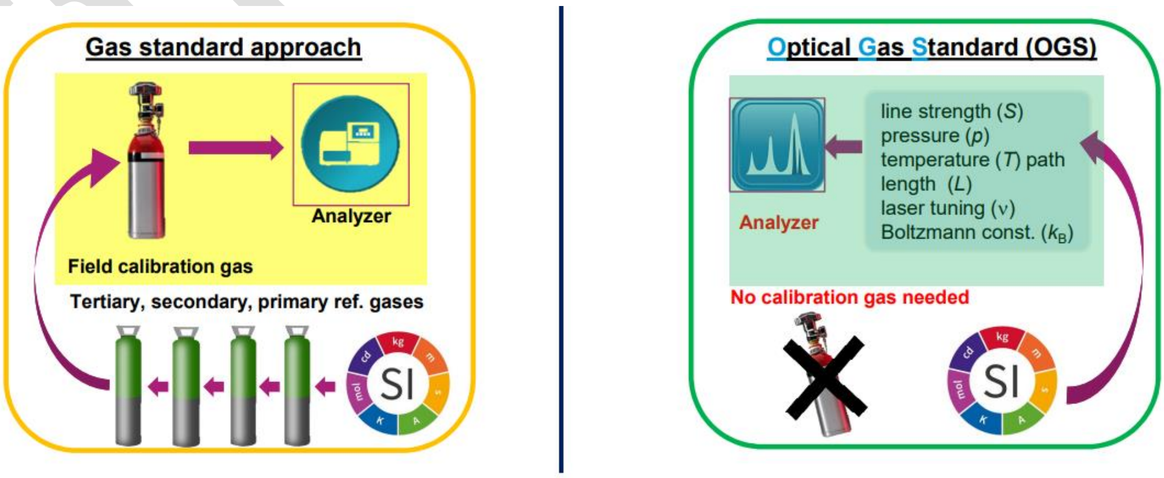
Metrology is the science of measurement, ensuring comparability, reliability and accuracy of measurement results (De Bièvre 2012). Metrological traceability is a central concept in metrology, providing the link between the measurement result and the standard (i.e., a primary realization of the unit of the measurement, typically the SI system of units) through an unbroken chain of calibrations. Metrology has been serving ambient ammonia measurements through gaseous calibration standards (van der Veen 2012), and standard measurement techniques (EN 17346:2020) (European Standard 2020). The recent EMRP project "Metrology for ammonia in ambient air" (Pogány et al. 2016) aimed to strengthen the link between the gas metrology community and field measurements. The main objectives of the project included 1) improving the quality of static and dynamic reference gas standards, 2) improving the accuracy of spectroscopic measurement techniques and 3) organizing extensive inter-comparison studies. Within this project, our aim was to convert a commercial cavity ring-down spectrometer into an instrumental optical gas standard (OGS, see Figure 13), i.e., a spectrometer capable of absolute and traceable ammonia concentration measurements without the need for calibration using gaseous standards. Besides the existing reference gas standards, this approach brings further possibilities to the operation, calibration and validation of field instrumentation. An OGS can be used to calibrate or validate other instruments using gas mixtures or ambient air, and can also be used for in-situ calibration-free gas analysis. In both cases metrological traceability is provided by the traceability of the OGS, replacing the need for traceable calibration gas mixtures. The OGS concept has been realized and demonstrated for laser-based instruments detecting H_2O (Buchholz et al. 2014), CO, NO_2, as well as HCl, the latter recently validated in a direct metrological comparison (EURAMET 2021).

Figure 13: Calibration strategies using gaseous reference standards (left) vs. an instrumental optical gas standard (right).



Source: The EMRP project "Metrology for ammonia in ambient air"

In this study we used a Picarro G2103 cavity ring-down spectrometer. The spectrometer hardware was combined with a device-external, proprietary, first principles spectral data evaluation to achieve absolute concentration measurements. Our data evaluation takes into account all relevant spectral lines of NH_3, H_2O and CO_2 in the detected spectral window using a least-squares solver by the Danish Fundamental Metrology (DFM) institute (Nielsen 2001). NH_3 concentration[7] is calculated from the integrated NH_3 line area determined from the measured absorption spectra using the Beer-Lambert law, high accuracy line intensities of the NH_3 lines (which were measured at PTB) and measured physical properties of the gas sample. By comparing to traceable reference sensors, we found that both the temperature and pressure sensors of the spectrometer are accurate within 1 %. The wavenumber axis of the measured spectra was validated by comparing the observed positions of the absorption lines to the values listed in spectroscopic databases, deviations were found to be below 1 %. We examined temperature sensitivity of the spectrometer and the gas handling system in a climate chamber. Rapid temperature changes in the range of 10 to 30 °C ambient temperature had no measurable effect on the derived NH_3 concentrations. Response time of the spectrometer was found to be in the range of minutes (10-90 % < 1.5 minutes, 1-99 % < 30 minutes in case of steps of 50, 100 and 200 ppb[8] in both directions). Cross-sensitivity to CO_2 was found to be negligible: a step from 0 to 440 ppm CO_2 in the gas sample led to less than 0.25 % relative change in the measured NH_3 concentration. H_2O cross-sensitivity was found to be minor: a step from 0 to 10000 ppm H_2O concentration generated only 2.4 % relative change in the measured NH_3 concentration. Although these cross-sensitivities are smaller than the uncertainty of the NH_3 concentration measurements, they might be of importance in high-precision measurements or in case of extreme H_2O or CO_2 concentrations.

Table 6: Results of the field comparison of the optical gas standard (OGS) and a mobile reference gas generator (ReGaS) (Twigg et al. 2022).

Concentration provided by the ReGaS generator /ppb	Expanded uncertainty (k=2) /ppb	Concentration measured by the OGS /ppb	Expanded uncertainty (k=2) /ppb	Normalized error
0	n/a	0.14	0.62	0.23
9.93	0.18	9.72	0.66	0.31
24.51	0.43	24.09	0.92	0.41
39.95	0.69	41.09	1.30	0.77

The OGS was compared to a traceable dynamic reference gas generator, *ReGaS* based on a permeation source, which was developed by METAS, the Swiss national metrology institute (Pascale et al., 2017). We performed the comparison in the field, inside an air-conditioned mobile laboratory and found good agreement between the two standards (see Table 6, we note that more data are reported in Twigg et al. 2022), here we report only those, where a sufficient stabilization time was given). Relative expanded uncertainties of the concentrations measured by the optical gas standard were in the range of 2-6 %, comparable to that of the ReGaS (< 3 %). In Table 6, we included the so-called normalized error, being a common measure of agreement between measurement results. If the normalized error is below 1, the two results are metrologically

[7] *amount fraction* in metrology
[8] ppb and ppm: the corresponding SI units are nmol/mol and µmol/mol, respectively

compatible, i.e. the difference between them is smaller than the uncertainty of this difference. As we see in the table, the normalized error is considerably smaller than 1 indicating good agreement between the results at all measured concentrations. We note that the applied reference gas generator ReGaS is also a new development, completely independent from our OGS, thus these results promote confidence in both standards.

Finally, our OGS participated in a field measurement campaign, together with 12 other instruments including wet chemical analyzers and several spectroscopic instruments (Twigg et al. 2022). No technical issues occurred with the OGS throughout the field campaign. Comparing the OGS to the different NH_3 instruments at various ambient concentrations revealed relative deviations of up to 20 %. More details on the possible origin of these deviations including the influence of different instrument locations, sampling line designs and states, as well as different time resolutions can be found in Twigg et al. (2022).

In summary the performance of our OGS was found to be suitable for calibration and validation experiments and its applicability under field conditions has been demonstrated. Further work is planned to elaborate a detailed operation protocol including critical maintenance of the sampling line and filters to minimize sampling artifacts and to ensure reliable and comparable NH_3 measurements in ambient air.

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4.1.11 Long-term NH_3 measurements at the TROPOS research station Melpitz using a MARGA

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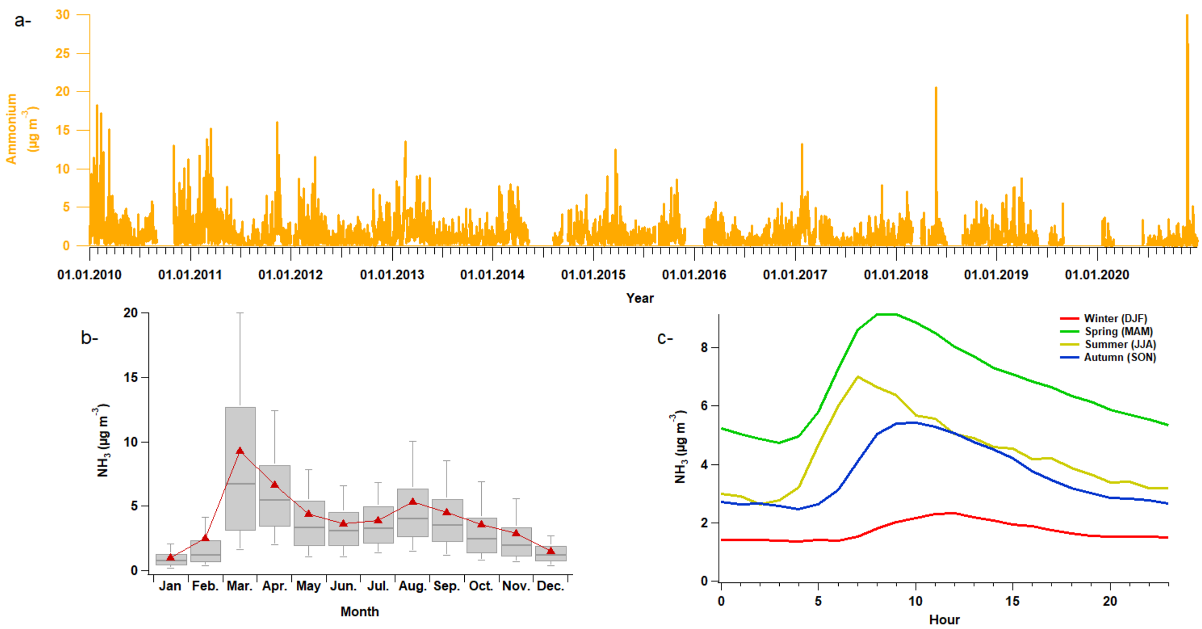
Since 2010, an hourly quantification of inorganic water-soluble PM_{10} ions and corresponding water-soluble trace gases including NH_3 is performed using a Monitor for AeRosols and Gases in ambient Air (MARGA) at the TROPOS research station Melpitz (12°56′E, 51°32′N, 86 m a.s.l.), located about 50 km to the northeast of Leipzig, Germany. The station has been in operation since 1992 to examine the impact of atmospheric long-range transport on central European background air quality (Spindler et al. 2012; Spindler et al. 2013). The site itself is situated on a meadow and is mainly surrounded by agricultural pastures and forests. Melpitz station is part of EMEP (European Monitoring and Evaluation Programme; Level 3 station; (Aas et al. 2012)), ACTRIS (Aerosols, Clouds and Trace Gases Research Infrastructure), GAW (Global Atmosphere Watch of the World Meteorological Organization), and GUAN (German Ultrafine Aerosol Network; (Birmili et al. 2009)). During several months, ammonia measurements from MARGA were compared with offline (NH_3 mini denuder, Midefix, Radiello), as well as online (PICARRO G1103) systems (Stieger et al. 2018). This unique dataset provides us the opportunity to better understand the dynamics of ammonia over years (Figure 14a), months (Figure 14b), seasons, and during the day (Figure 14c).

Figure 14: Time series of the hourly ammonia concentration measured by a MAGRA over 10 years (a), the corresponding monthly average (b), and the seasonal diurnal patterns (c)



Source: TROPOS, Leipzig.

The MARGA deployed at Melpitz was further developed and combined with an additional ion chromatography (Compact IC) to allow quantifying low molecular-weight organic acids (such as formic, acetic, propionic, butyric, pyruvic, glycolic, oxalic, malonic, succinic, malic, glutaric, and methanesulfonic acid) in both gas and particle phases with a 2-hour time resolution (Stieger et al. 2019). Measurements were used to investigate the partitioning of the low-molecular-weight organic acids. The results show a strong deviation of the thermodynamically expected phase partitioning of the measured low-molecular-weight organic acids (Stieger et al. 2021). Recent work emphasized that the reaction between organic acids and ammonia can lead to the formation of stable organic salts (e.g. (Schlag et al. 2017). For this reason and regarding the availability of ammonia at Melpitz, ammonia might have a significant role in the observed particle-phase enrichment by inducing the formation of organic salts species. As a consequence, it appears that ammonia is not only a key species for the formation of the secondary inorganic species like ammonium sulfate and ammonium nitrate, but it could also directly impact the organic aerosol mass concentration.

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4.1.12 NADP's ammonia monitoring network (AmoN): Long-term measurements to support deposition and critical load assessments in the United States

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Since 1978, the National Atmospheric Deposition Program (NADP) has tracked the status and changes in wet deposition within precipitation across the continent. Beginning in 2010, the NADP operated the Ammonia Monitoring Network (AMoN) to provide data to evaluate long-term trends in ambient ammonia concentrations and deposition. Currently, there are 116 AMoN sites across the U.S. and Canada, and over 40,000 gaseous observations of atmospheric ammonia in our publicly available database.

This talk provided details on the operation of the AMoN, including methods, available observations, analytical results along with quality assurance information, and annual temporal and spatial NH_3 concentrations over the 10-year data record. The goal was to allow for comparison to equivalent networks across Europe to improve the global monitoring and modeling assessments. Additionally, we will provide specifics from several efforts that use this ammonia data, including US E.P.A.'s Clean Air Status and Trends Network (CASTNet) and the U.S. National Park Services (NPS). EPA uses their CASTNet observations and the AMoN results to estimate dry deposition of nitrogen species through a measurement-model fusion process (CMAQ, Community Multiscale Air Quality Modeling System). The NPS uses these wet and dry deposition estimates of nitrogen, in collaboration with the NADP's Critical Loads Atmospheric Deposition (CLAD) committee, to estimate total deposition of nitrogen in Rocky Mountain National Park (Colorado), as a comparison to critical loading of N in the Park, and to help determine the sources of this excess nitrogen.

Further Reading:

EPA Measurement-Model Fusion Dry Deposition Results:

https://nadp.slh.wisc.edu/committees/tdep/

NADP's Total Deposition Mapping utility:

https://clmapper.epa.gov/

Rocky Mountain Nitrogen Initiative:

https://cdphe.colorado.gov/public-information/planning-and-outreach/rocky-mountain-national-park-initiative

https://cdphe.colorado.gov/public-information/planning-and-outreach/rocky-mountain-national-park-initiative/monitoring-and

4.2 Workshop Day 2, 29 March 2022

4.2.1 Relationships between NH_3 in urban air and the diversity of epiphytic macrolichens

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Emissions of nitrogen oxides (NO_x) from traffic are implicated in changing the diversity of epiphytic lichens with a shift to nitrophytes (Davies et al. 2017). The increasing use of three-way catalysts in cars (Borsari and de Assuncâo 2017) and Selective Catalytic Reduction systems in diesel vehicles (Stelwagen and Ligterink 2015) reduces the emissions of NO_x but increases ammonia (NH_3) emissions (Reche et al. 2012).

NH_3 is harmful especially to acidophytic lichens (Wolseley et al. 2009). We studied the relationship between ambient NH_3 concentration and the diversity of epiphytic lichens in Helsinki (60°10'N, 24°56'E), southern Finland, to verify the results of a previous study by Manninen (2018) which suggested that NH_3 emissions from vehicles in the area may be high enough to affect lichens.

Epiphytic macrolichens were scored on *Acer platanoides*, *Ulmus glabra*, *Quercus robur*, and *Pinus sylvestris* trunks at 20 sites in Jul-Oct 2019 based on the European Standard EN 16413:2014 (Finnish Standards Association 2014). Bark samples were collected for pH measurements and lichen thalli for analysis of total N concentration. Monthly NH_3 concentrations were measured for two years (Oct 2019 - Sep 2021) with passive samplers (ALPHA®, Tang et al. 2001). Modelled NO_2 concentrations in 2019 were taken from a concentration map provided by the Helsinki Region Environmental Services Authority (https://www.hsy.fi/en/air-quality-and-climate/air-quality-now/annualairqualitymap/). Lichens species with eutrophication scores of 1-4 were classed as acidophytes and those with 5-9 as nitrophytes (Wirth 2010) for calculating a Lichen Diversity Value separately for each tree species per site (LDV). Corresponding values for acidophytes (LDV_A) and nitrophytes (LDV_N) were also calculated (Finnish Standards Association 2014) as was the Lichen Atmospheric Nitrogen index (L_{AN}) (Wolseley et al. 2019).

The monthly NH_3 concentrations ranged from 0.00 to 3.29 $\mu g\ m^{-3}$ and the 2-year mean concentrations from 0.15 to 1.03 $\mu g\ m^{-3}$. The average bark pH values were: *Pinus* 3.30, *Quercus* 4.84, *Acer* 5.35, *Ulmus* 5.65. The acidophytic *Hypogymnia phyosodes* was the most abundant species on *Pinus*, while nitrophytic *Parmelia sulcata* (*Quercus*) and *Physcia tenella* (*Acer, Ulmus*) had the highest abundances on deciduous trees across the sites. The abundance of *H. physodes* and the LDV_A were negatively correlated with NH_3 concentration, NO_2 concentration, and bark pH across deciduous trees, while the L_{AN} on *Quercus* and across deciduous trees increased both with increasing NH_3 concentration and bark pH. At the tree species level, the diversity of macrolichens on *Quercus* seemed to be the best indicator of NH_3. Overall, our results showed a decrease in the abundance of acidophytes and an increase in that of nitrophytes at NH_3 concentration below the current critical level of 1 $\mu g\ m^{-3}\ yr^{-1}$ (Cape et al. 2009). The impact of traffic-derived NH_3 emissions also appeared as an increase in the total N concentration of *P. sulcata* with increasing NH_3 concentration.

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4.2.2 Results from an ammonia fumigation and a field study using endangered nitrogen-sensitive plant species

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Ammonia (NH_3) acts as a plant nutrient and leads to eutrophication and acidification in N-sensitive ecosystems. Research on the long-term effect of low NH_3 concentrations on higher plant species is sparse. Natural gradient studies are focussing on lower plants such as lichens and an NH_3 field fumigation system has solely been used at Whim Bog, Scotland. With a greenhouse fumigation and a field gradient approach, we aimed to provide new data for the discussion of critical levels (CLe).

Greenhouse study 2020

Our first approach was a greenhouse study conducted in 2020 in Stuttgart Hohenheim (Germany). Diluted ammonia solutions were pumped into four greenhouse chambers with a peristaltic pump where one chamber acted as a control without fumigation. The solutions were allowed to drip into plastic containers from which they gently vaporized. The vaporizing gas was distributed with fans in each chamber and the air ammonia concentrations were measured weekly with Radiello® passive samplers. Additionally, temperature and humidity were determined and the four treatments were rotated between chambers in order to minimize chamber effects. Seven plant species were chosen according to a low Ellenberg N value and their threat of extinction (*Antennaria dioica*, *Arnica montana* with two different seed origins, *Carex arenaria*, *Dianthus deltoides*, *Koeleria glauca*, *Molinia caerulea* and *Pulsatilla vulgaris*). 20 plants (one per pot) were then exposed in the chambers starting on 30 April 2020.

Watering was conducted manually with rain water and half of the pots were additionally undergoing drought stress, i.e. they were receiving around one third less water. During the season, parameters like the plant length and flower number were determined until a species-individual intermediate harvest and a final harvest was conducted at the end of the season. The aboveground biomass was cut, dried and weighed for each plant. Each species was exposed for an individual duration of time, until the advent of senescence in most plants. Temperatures over the season and throughout days did not differ much between the chambers but were generally high, reaching daily maxima of over 40 °C. Measured ammonia concentrations ranged between 4 and 18 µg m^{-3} averaged over the season. The three lowest concentrations were close (4-5 µg m^{-3}) to each other and the two highest concentrations were around 8 and 18 µg m^{-3}. With regard to the dry treatments, all species produced less biomass in at least one harvest. Only *P. vulgaris* without intermediate harvest were not affected by the drought, corresponding to the plant's xerophytic properties.

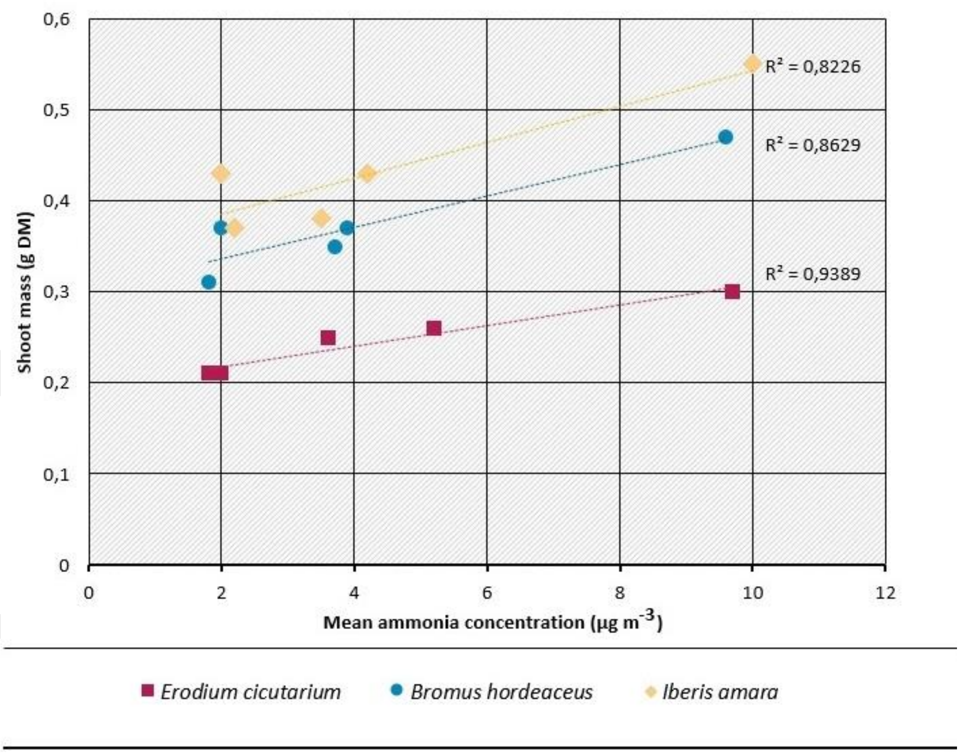
When looking at the different ammonia concentrations, plant reactions differed from species to species. Whereas most species revealed no clear reaction pattern, the grass *M. caerulea* showed a slight trend to increase biomass production in the well-watered plants. In the dry treatments, the opposite trend could be observed. A reason for the decreased growth could be that the higher level of NH_3 could not be assimilated by the stressed plants due to more frequent stomata closure.
Summarizing the greenhouse study, we found the fumigation set-up worked well, but planned NH_3 concentrations below 10 µg m^{-3} could not be realized and did not approach zero in the control. Furthermore, high temperatures hindered plant development, i.e. flower production in many species. With regard to the dry treatment, most species reacted similarly, with a reduced biomass

production, whereas the reaction to the different ammonia treatments varied substantially not only between species but also within the same species depending on the harvests and watering (example of *M. caerulea*).

Field study 2021

The second experiment was a field gradient study conducted at a University farm Unterer Lindenhof (ULI) in the lee of livestock stables. All five stations were chosen after a NH_3 dispersal model and at all locations, NH_3, temperature, humidity and wind parameters were measured. Furthermore, we determined the rain quantity and chemistry (ammonium and nitrate) at all stations. Plants from the greenhouse experiment as well as new species were added. These were one perennial with 20 plants per treatment, *Dianthus gratianopolitanus* and five annuals with 10 plants per treatment, *Bromus hordeaceus*, *Erodium cicutarium*, *Iberis amara*, *Legousia speculum-veneris* and *Trifolium arvense*. Plants were exposed in pots on metal tables. Due to the rather moist conditions over most of the season 2021, no drought treatment could be realized. In contrast to the previous year, no intermediate harvest was performed. For the data analyses, we still distinguished between well-watered and dry-treated plants and plants with/without intermediate harvest to investigate potential differences in growth and carry-over effects from the previous season. Measured temperatures at the five stations were rather cool throughout the season and did not differ between the stations. The weekly determined NH_3 concentrations showed a gradient between 2 and 10 µg m^{-3} averaged over the season. Station 3 revealed slightly higher concentrations than station 4, not corresponding to the underlying dispersal model.

Figure 15: Shoot dry weight (g) of *E. cicutarium*, *B. hordeaceus* and *I. amara*



Source: Own illustration, University of Hohenheim.

Plants that had already been exposed to ammonia in the previous season, were generally producing more biomass in 2021 when no intermediate harvest was performed in 2020. On the other side, nearly no carry-over effects with regard to biomass production could be observed. With regard to the different ammonia concentrations, species varied in their reactions. Several species showed no differences between the five stations, while others produced less biomass under higher NH_3. *M. caerulea* again showed an increased growth, but this time only the last year's dry treated plants produced more biomass at higher levels of NH_3. A reason might be that plants which produced less biomass in the previous season saved more nutrients that could be used for an increased growth in the second season.

Regarding the newly added species, we found that the perennial *D. gratianopolitanus* did not react to the different ammonia concentrations, but three out of the five annuals produced more biomass. *E. cicutarium*, *B. hordeaceus* and *I. amara* responded linearly to increasing levels of NH_3 (R^2 of 0.94, 0.88 and 0.82, respectively, see Figure 15). The leguminous annual *T. arvense* on the contrary tended to produce less biomass with increasing NH_3 concentrations (R^2 0.68) while *L. speculum-veneris* revealed no differences between the stations.

Summary

All in all, tested plant species responded differently to the different ammonia concentrations. In both seasons, some species were not affected, while others produced more or even less biomass under higher ammonia and even within one and the same species reactions could differ. We saw that NH_3 was more beneficial in annual species and that it hindered growth in other species, especially the tested legume. Therefore, we hypothesize that legumes will be more sensitive towards ammonia, a question that should be addressed in further ammonia fumigation experiments. It remains to be discussed whether only ammonia affected the plants in the field or if stations close to the pig stable might have been affected by other gaseous compounds and/or (N-containing) particles. However, the bulk deposition at the five stations showed no differences. Based on our experiments, gradient studies with pre-grown plants are preferred over greenhouse/chamber studies and we call for further ammonia studies in order to address other plant species for the refining of critical levels in the future.

4.2.3 Towards understanding shorter-term critical levels with monthly resolution measurements

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Elevated atmospheric ammonia (NH_3) concentrations and nitrogen (N) deposition pose a substantial threat to N-sensitive vegetation in Northern Ireland (NI). Large areas of NI experience annual mean NH_3 concentrations above the critical levels, with 80% of N-sensitive habitats exposed to concentrations >1 µg NH_3 m^{-3}, and 14% of N-sensitive habitats > 3 µg NH_3 m^{-3}.

Ammonia monitoring is vital for verifying national modelled annual estimates of NH_3 concentrations, to quantify the level of threat to designated sites and for understanding local sources and their spatial and temporal patterns. For the study described here, local atmospheric ammonia monitoring networks were established for eight sites of European Importance (Special Areas of Conservation, SAC), combined with vegetation assessment, including vegetation N content. Atmospheric NH_3 has been measured at monthly intervals using triplicate passive ALPHA® samplers at the following designated sites in NI, with 1-9 sampling sites per designated site (all SACs are in lowland areas surrounded by agricultural activities of different levels of intensity, except for who upland sites, as specified below):

- Ballynahone Bog (since September 2014)
- Other sites (since June 2020)
  - Curran Bog
  - Garry Bog
  - Moneygal Bog
  - Peatlands Park
  - Slieve Beagh (upland site)
  - Turmennan
  - Cuilcagh Mountain (upland site)

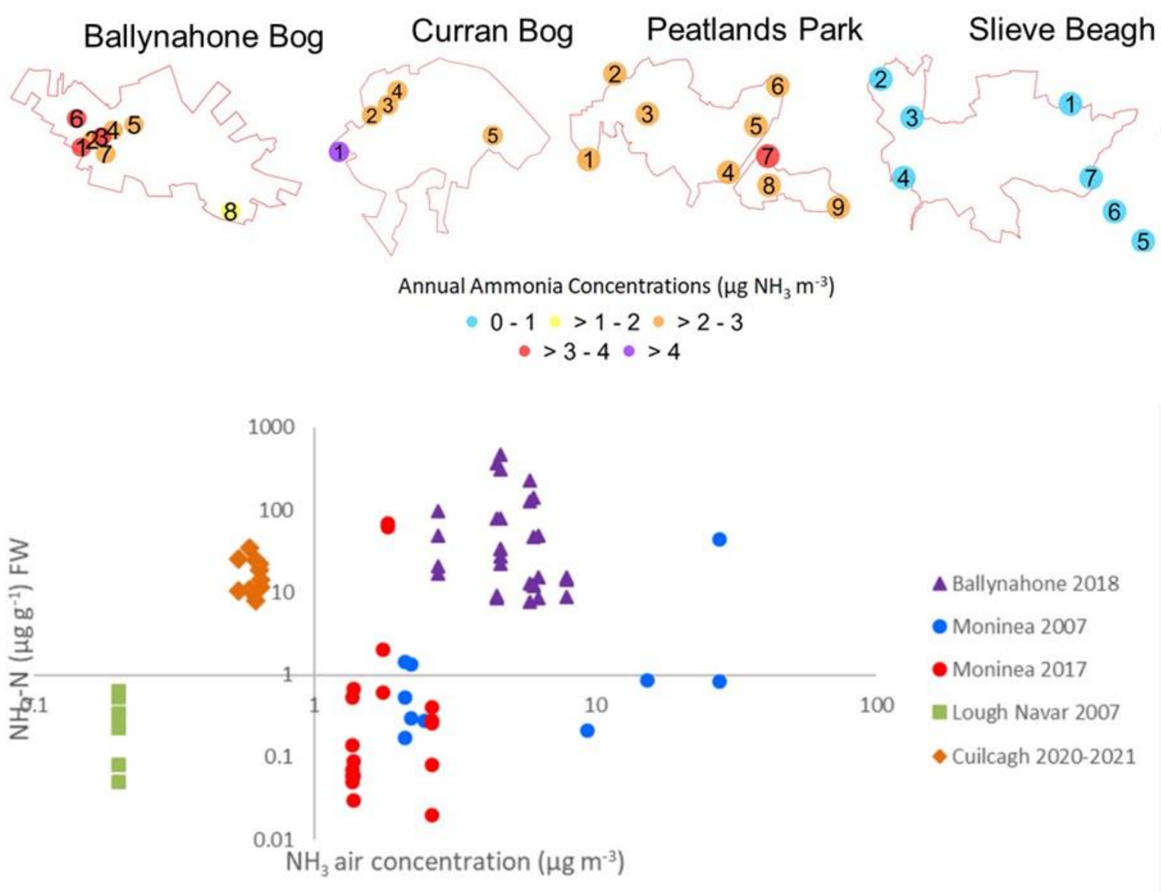
All sampling sites except for the two upland sites, Slieve Beagh and Cuilcagh Mountain, exceed the annual 1 µg NH_3 m^{-3} critical level, and several monitoring sites exceed the 3 µg NH_3 m^{-3} critical level across the lowland sites (four examples shown in Figure 16, top). Monthly measurements show high temporal variability, both within-year and between years, the latter highlighting, for example, different wind patterns which influence the relative importance of prevailing vs. less frequent wind directions, as well as wind speed determining the build-up or dispersion of plumes.

In line with agricultural activities, much higher NH_3 concentrations are normally observed in Northern Ireland during spring in particular (with a smaller secondary peak in early autumn), when slurry and manure applications occur. Therefore, by focusing on average annual concentrations, the underlying seasonal peaks due to local activities in the vicinity of the designated sites are often not considered in modelling assessments, which do not have a sub-annual temporal resolution. Local hotspots and gradients away from sources into designated sites are also underestimated with UK national scale modelling which quantifies grid square average concentrations that smooths out the high spatio-temporal variability at the field and farm scale which is the relevant scale for assessing local impact.

Vegetation N content (Figure 16, bottom) from five sites in Northern Ireland (including two of the above sites) has been assessed using foliar ammonium (NH_4) content (and % N for a subset), from samples taken in February (across different years). In most cases, the foliar NH_4-N concentrations correlate well with atmospheric NH_3 concentrations, at the lowland SACs embedded in agricultural land, and can be explained as due to local intensive sources within wider agricultural activities taking place. The exception is the upland site (Cuilcagh Mountain), where relatively low atmospheric NH_3 concentrations, averaging at 0.6 µg NH_3 m^{-3} over the year (0.1-1.2 µg NH_3 m^{-3} monthly averages) coincide with relatively high NH_4 in the vegetation. This is expected to be due to elevated N deposition in this remote upland landscape, and a first year of wet deposition sampling is currently in progress.

Figure 16: Average annual NH_3 concentrations at four designated nature conservation sites in Northern Ireland (top) and vegetation N content (foliar ammonium) at four sites in Northern Ireland (bottom).

Moninea Bog was sampled twice, in 2007 while a poultry farm next to the bog was operational, and in 2017, seven years after the farm had ceased operation



Source: Own illustration, CEH Edinburgh.

In conclusion, it is important to understand the temporal variability of atmospheric NH_3 and the potential impact of short-term peak concentrations, in addition to the annual average values used for comparison against critical levels. It is further important to consider the large concentration gradients away from local hotspots when assessing the impact of emission sources on sensitive species, habitats and designated sites. And finally, impacts from atmospheric nitrogen on vegetation are a complex combination of concentration-based effects (through elevated NH_3) and deposition-based impacts, in particular wet deposition at high-precipitation sites that may be relatively remote from local emission sources.

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Contributors

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4.2.4 Biomonitoring of ammonia with the epiphytic lichen *Hypogymnia physodes*

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Abstract

The usage of epiphytic lichens as bioindicator has a long tradition in biomonitoring of air pollution. The epiphytic lichen *Hypogymnia physodes*, which was very common in the past, is relatively insensitive to acidic air. This lichen species has recently been declining significantly in many rural regions of Central Europe as a result of ammonia input, decreasing precipitation acidity and climatic changes. Between 1998 and 2020, a significant decline of *Hypogymnia physodes* was observed in a Northwest German pine forest (Sandkrug). Within this period, mean ammonia concentrations varied between 2 µg m^{-3} and 3 µg m^{-3} and N throughfall deposition dropped from about 30 kg ha^{-1} yr^{-1} to about 20 kg yr^{-1} a^{-1}. At the same time, the pH of pine bark increased slightly from 4.3 to 4.5. Lichen thalli exposed to different atmospheric NH_3 concentrations higher than 2.0 µg m^{-3} in a forest clearing of Sandkrug and in the Botanic Garden of Oldenburg between 2019 and 2021 showed clear injuries already after 12 months. In the following months, these injuries intensified with significant differences. Mean NH_3 concentrations of 1.5 µg m^{-3} led to a moderate injury of the lichen thalli.

The studies in northwestern Germany confirm the high impact of enhanced NH_3 air pollution on *Hypogymnia physodes* and other NH_3-sensitive lichen species and provides the possibility of NH_3-specific biomonitoring by means of mapping.

Introduction

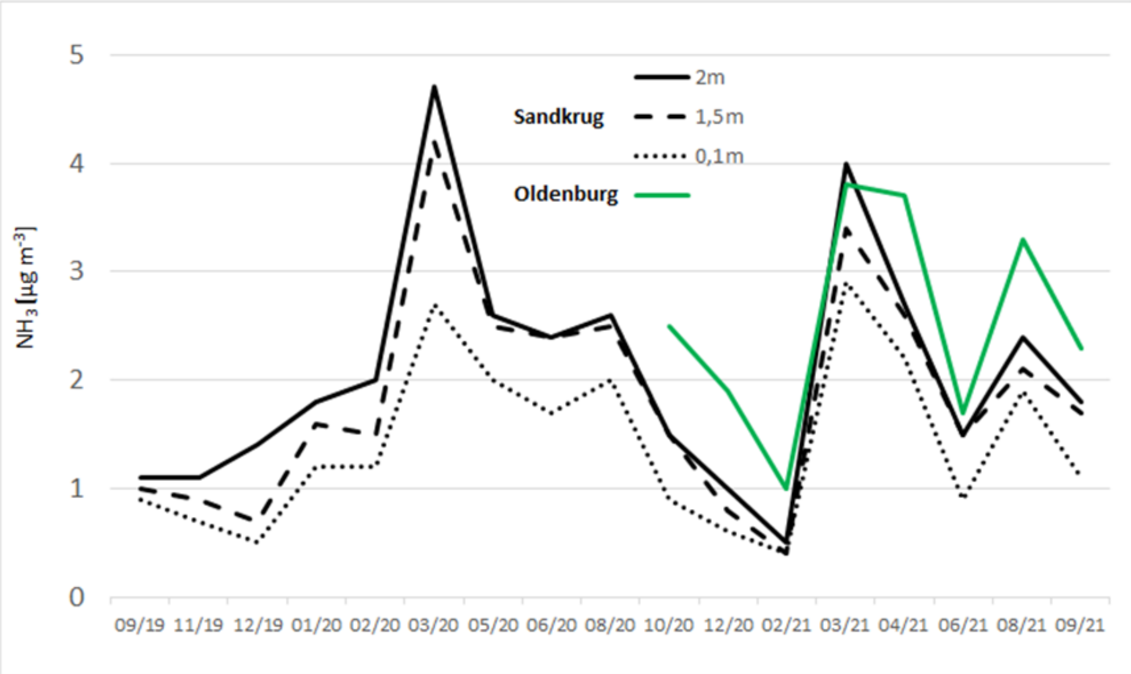
The usage of epiphytic lichens as bioindicators has a long tradition in biomonitoring of air pollution because they react very sensitive to air pollutants. The epiphytic lichen *Hypogymnia physodes*, which was very common in the past, is relatively insensitive to acidic air pollution (Werner 1993) and has recently been declining significantly in many rural regions of Central Europe. Enhanced air pollution of ammonia is considered to be the main driver for this decrease (deBryun et al 2009, Kolk et al. 2020, Wolseley et. al 2004). In Northwest Germany, exposure experiments were carried out to get more detailed information about the sensitivity of this lichen species towards ammonia (Mohr 2021). This paper shows further results of these experiments.

Study area

The investigations in Northwestern Germany were carried out in intensively used agricultural areas and adjacent areas in the federal state of Lower Saxony where ammonia plays an important role as air pollutant. In open land, the level of NH_3 concentrations in the air is about 3 µg m^3 to 6 µg m^3. Higher NH_3 concentrations are measured in the vicinity of farms, which led to a die back of exposed *Hypogymnia physodes* in a short time (Mohr 2001).

For the exposure experiments with *Hypogymnia physodes* under lower air pollution conditions the pine forest "Sandkrug" as well as the Botanical Garden in the city of "Oldenburg" were chosen. In Sandkrug, ammonia air concentrations were measured in a clearing, as well as N throughfall and bark pH were measured in pine stands since 1998. Since then, the yearly mean NH_3 concentration decreased from 3 µg m^{-3} to 2 µg m^{-3} and the N throughfall deposition decreased from about 30 kg ha^{-1} yr^{-1} to about 20 kg yr^{-1} a^{-1}. The pH of pine bark initially colonized with lichens increased slightly from 4.3 to 4.5 during this period. During the exposure periods, the 4-weekly to 6-weekly NH_3 concentrations in 2 m height varied between 1.0 µg m^{-3} and 4.7 µg m^{-3} in Sandkrug and and between 1.0 µg m^{-3} and 3.8 µg m^{-3} in Oldenburg (Figure 17).

Figure 17: NH_3 concentrations (4 – 6 monthly average) in Sandkrug (3 heights: 2m, 1.5m, 0.2m) and in Oldenburg (2m)



Source: Own Illustration, Landwirtschaftskammer Niedersachsen, Oldenburg.

Method

Following VDI (2019), vital lichen thalli of *Hypogymnia physodes* from low polluted regions in Eastern Germany were fixed on a wooden board and exposed up to 2 years, until most of the lichens showed substantial injuries. In Sandkrug, several boards were exposed in different heights, which generally leads to different NH_3 air concentrations due to NH_3 absorption of the ground vegetation (Figure 18). An influence of more favorable growth conditions at lower exposure heights could not be confirmed at natural sites of *Hypogymnia physodes*. Additionally, the equally common but less NH_3-sensitive species *Parmelia sulcata* was exposed for comparison purposes. For evaluation, the lichen thalli were photographed at one- to several-month intervals and the degree of damage was documented in 10% increments. Simultaneously, NH_3 concentrations were measured using IVL passive samplers. More detailed information about the study methods is given in Mohr (2021).

Results

Hypogymnia physodes was very common on acid barks of trees in Sandkrug and other regions in Northwest Germany until the 1990s. Subsequently, a strong decline of *Hypogymnia physodes* and other acidophytic species (e.g. *Platismatia glauca*, *Pseudevernia furfuracea, Tuckermanniopsis chlorophylla*, *Usnea ssp.*) was observed. Recently, only *Hypogymnia physodes* still exists sporadically on shrubs close to the ground in Sandkrug.

Thalli of *Hypogymnia physodes* sampled from remote areas were exposed on wooden panels in a clearing of the pine forest Sandkrug. After a period of 12 months, first damages were observed (5%

to 10% degree of injury, Figure 18. After a period of 2 years, the most damaged thalli were found at an exposure height of 2 m, where a mean NH_3 concentrations of 2.1 µg m^3 was measured in the exposure period.

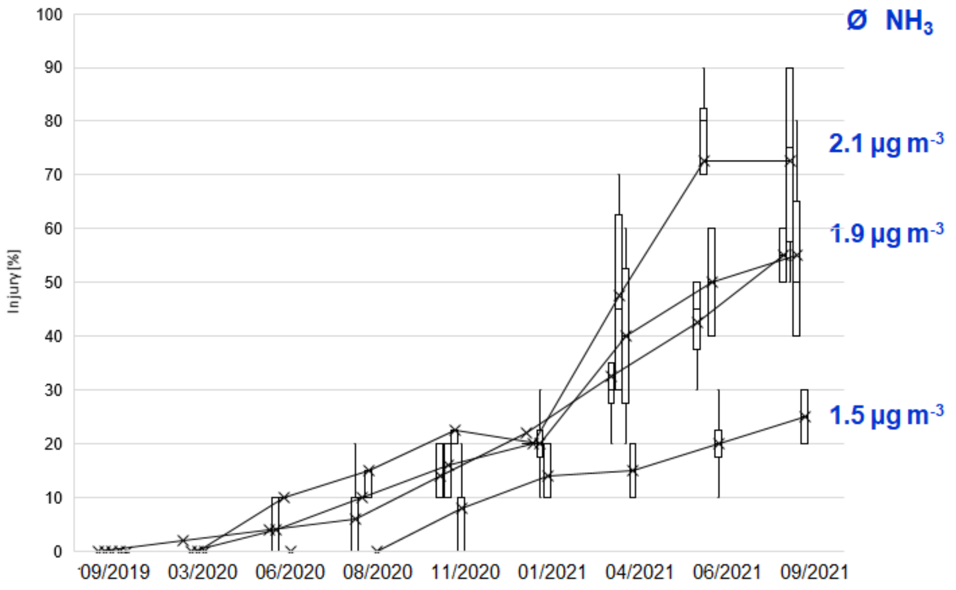
Figure 18: Exposure boards with *Hypogymnia physodes* in heights between 0.2 m and 2 m in Sandkrug

Source: Own photograph, Landwirtschaftskammer Niedersachsen, Oldenburg.

At the end of the period, almost all lichen thalli in 2 m had died off. At lower heights (1.5 m, 1 m) with mean NH_3 concentrations of 1.8 µg m^{-3}, the degree of injury was even on a high level between 50% and 60%. At the lowest exposure height (0.2 m) with mean NH_3 concentrations of 1.5 µg m^3, the injury of *H. physodes* reached 20% on average.

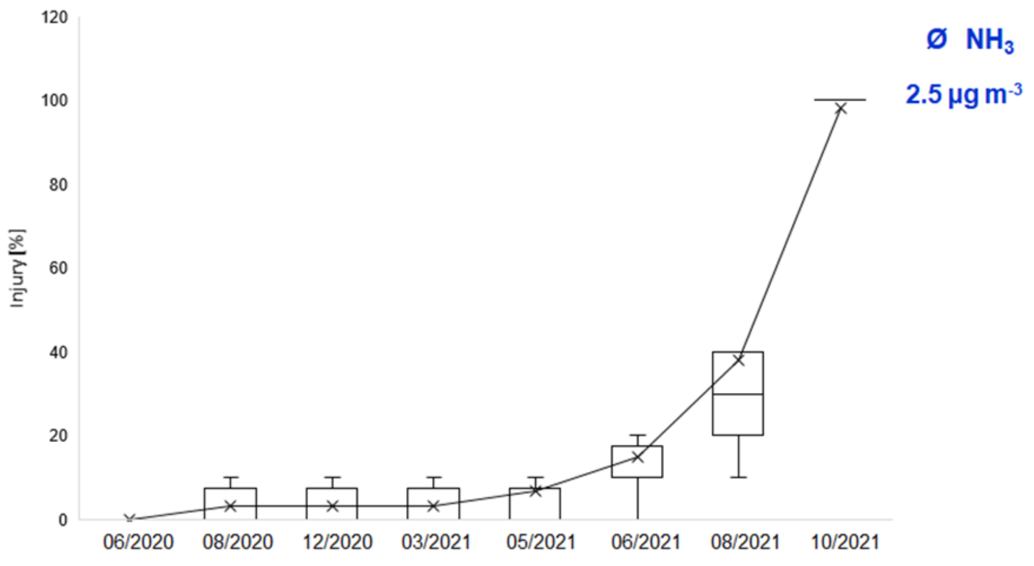
Figure 20 shows the progression of the degree of injury of the exposed lichens in Oldenburg, where mean NH_3 concentrations of 2.5 µg m^3 were determined. The first clear damage of *Hypogymnia physodes* occurred after 12 months. In the following 3 months, the lichen thalli declined successively and eventually disappeared. In contrast, the less ammonia sensitive species *Parmelia sulca* (Mohr 2001), which had been exposed on the same panel, remained largely undamaged (Figure 21).

Figure 19: Development of injury of *H. physodes* in Sandkrug within an exposure period of 2 years at different mean NH_3 concentrations (confidence interval, medium, average, min. and max. value of injury in %)



Source: Own illustration, Landwirtschaftskammer Niedersachsen, Oldenburg.

Figure 20: Development of injury (%) of *H. physodes* within 15 months in Oldenburg (confidence interval, medium, average, min. and max. value) at mean NH_3 concentrations of 2.5 µg m^{-3}



Source: Own illustration, Landwirtschaftskammer Niedersachsen, Oldenburg.

Discussion

Among epiphytic lichens, the species *Hypogymnia physodes* exhibits a relatively high sensitivity to ammonia. Although other environmental conditions, such as climate warming and decreasing precipitation acidity may also play a role, its large-scale decline is essentially the result of the large-scale increase in NH_3 pollution (Frahm 2007, Mohr 2021). The exposure experiments in Sandkrug and Oldenburg indicate that NH_3 concentrations as low as 1.5 µg m^3 lead to slight damage within 2 years. Due to the high tolerance against other pollutants and environmental effects, *Hypogymnia physodes* prove to be a suitable indicator of atmospheric ammonia pollution. Thereby, mapping of *Hypogymnia physodes* and other similarly NH_3 sensitive species (e.g. *Tuckermanniopsis chlorophylla, Hypocenomyce scalaris, Platismatia glauca, Pseudevernia furfuracea, Usnea ssp.*) can provide information about the spatial distribution of atmospheric ammonia.

Figure 21: Thalli of *Hypogymnia physodes* (left) and *Parmelia sulcata* (right) before (above) and after an exposure period of 15 months in Oldenburg

Source: Own photograph, Landwirtschaftskammer Niedersachsen, Oldenburg.

Considering the unfavorable growth conditions at the exposure boards, the results of this study agree with Sutton et al. (2022), who found a decline of *Hypogymnia physodes* and other acidophytic species at NH_3 concentrations above 1.7 µg m^{-3} based on long-term observations in the UK. The protection of other rare foliose und beard species, which were often associated together with *Hypogymnia physodes* in the past, may require even lower NH_3 ambient concentrations.

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4.2.5 How can we assess the impact of project contributions of ammonia on Natura2000 sites?

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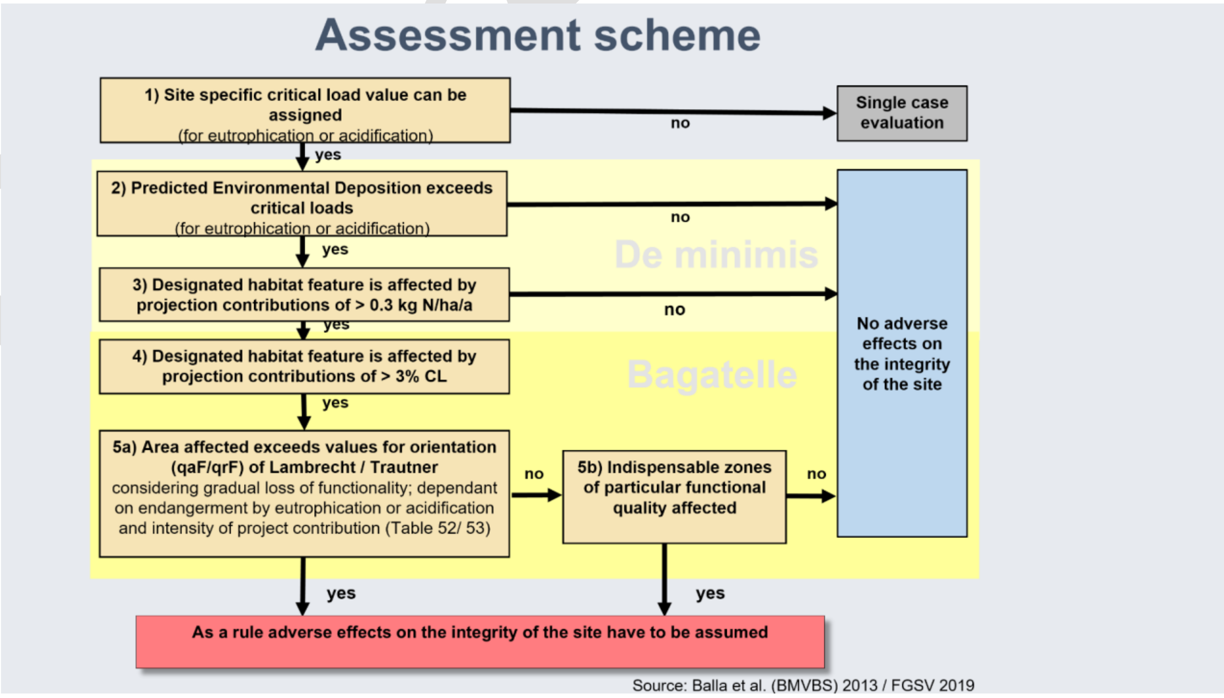
It is very common particularly for large infrastructure projects that they lie in or in the vicinity of Natura 2000 sites. In those cases, project contributions of nitrogen meanwhile are routinely considered within appropriate assessments under Article 6.3 of the Habitats Directive (HD). Only if adverse effects of the project on the affected site can be ruled out with certainty, a project may be permitted, otherwise an exception may only be granted under strict conditions.

Among the targets protected by the HD its Annex II lists a number of mosses. Also, lichens may be characteristic of habitats listed in annex II of the HD, thus also underlying the strict protection regime. Mosses and lichens, though, may be rather prone to damage by ammonia than by deposition of reactive nitrogen compounds in general. While different evaluation schemes for nitrogen deposition within appropriate assessments have been developed and established in the EU (Hicks et al. 2011, Whitfield & McIntosh 2014), little experience is available on the assessment of impacts by ammonia.

Figure 22 shows the assessment scheme established in Germany. To assess impacts by ammonia in a similar way, (1) critical loads may be substituted by critical levels and (2) values of background deposition may be substituted by values of ammonia concentrations in the air. Modelled values of ammonia concentrations are available at least in parts of Germany, notably in Baden-Württemberg (see Pruess, this issue). Less obvious is the answer to the next question: what might be an adequate *de minimis* criterion to distinguish relevant from irrelevant project contributions?

Figure 22: Assessment scheme of nitrogen deposition impacts within German appropriate assessments under the Habitats Directive



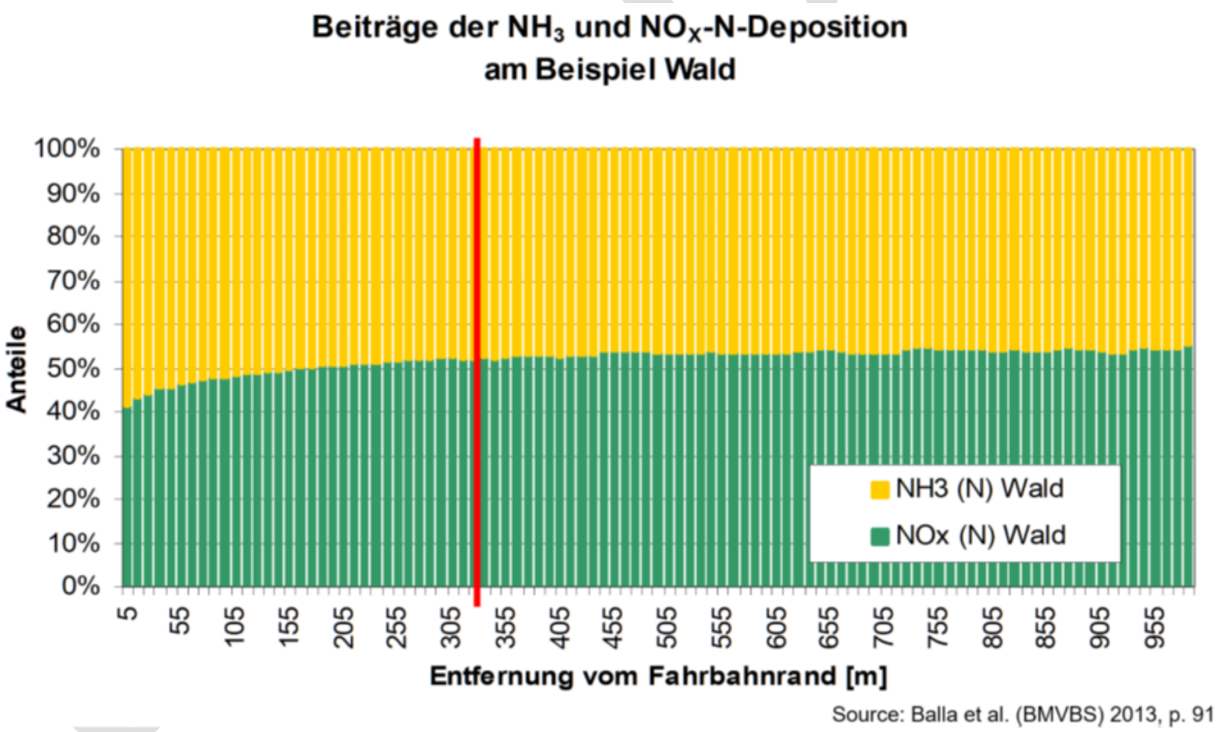
Source: Balla et al. (2013).

While there cannot be an exhaustive answer from the scientific point of view, it would be fallacious to think that there is a way around it: every site that is not subject to an assessment within a project permit procedure must have been found to be irrelevant by some kind of consideration, so at least implicitly a "*de minimis* value" is applied to rule out effects of nitrogen compounds.

The European Court has made very clear in numerous decisions that a precautionary approach has to be taken within appropriate assessments. Any project that may do harm to the targets of the HD, even single habitats within a protected site, must be carefully assessed. For this reason, *de minimis* criteria are much lower than the ones used under the air quality legislation.

Figure 23 shows the ratio of ammonia and NO_x depositions along a road. A typical distance where – depending on average traffic and particular emission rates – a *de minimis* criterion might apply is indicated by the red line at 330 m. Rather independent of the exact location of this line, fairly exactly half of the contributions (i.e. 50 %) may be attributed to ammonia. As the *de minimis* criterion has been set in Germany originally to assess the impacts of road projects, it makes sense to determine the amount of ammonia contained within the established *de minimis* criterion .3 kg N/ha/a also as half of this value, .15 kg N/ha/a; considering the span contained within the rounded value (.25 to just less than .35) it would amount to .1749 kg N/ha/a.

Figure 23: Ratio of road traffic contributions of ammonia- (yellow) and NOx-depositions (green) at different distances along a road (forest)



Source: Balla et al. (2013).

In order to (re)calculate air concentrations from depositions one has to consider deposition velocity and the purely mathematical transformation of spatial units (cm * ha vs. m^3) and temporal units (a to s). 1 $\mu g/m^3$ NH_3 calculates thus as v_D* 2.598 kg N/ha/a. With the deposition velocity that is typically applied in Germany for ammonia in forests set to 2 cm/s this results in a conversion factor of 5.2. An equivalent to the *de minimis* value of .3 kg N/ha/a to be applied for the effect of road projects would thus be 0.03 µg/m³ NH_3 (more exactly .03366 µg/m³); values of 0.04 µg/m³ or more would not be considered irrelevant.

This approach would give a *de minimis* value which is consistent with the well-established de minimis criterion for traffic borne project contributions of nitrogen deposition. More conventional work will be needed then, to agree on species specific critical levels and bagatelle values of project contributions to ammonia concentrations. In the absence of field experiments or existing expert values (both of which of course would be highly appreciated) an approach would be to assess it by careful observation of the conditions giving rise to a favourable conservation status of the species under consideration.

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4.2.6 Development of a trophic assessment and management system for habitat types (Directive 92/43/EEC) considering interrelations between critical levels and critical loads

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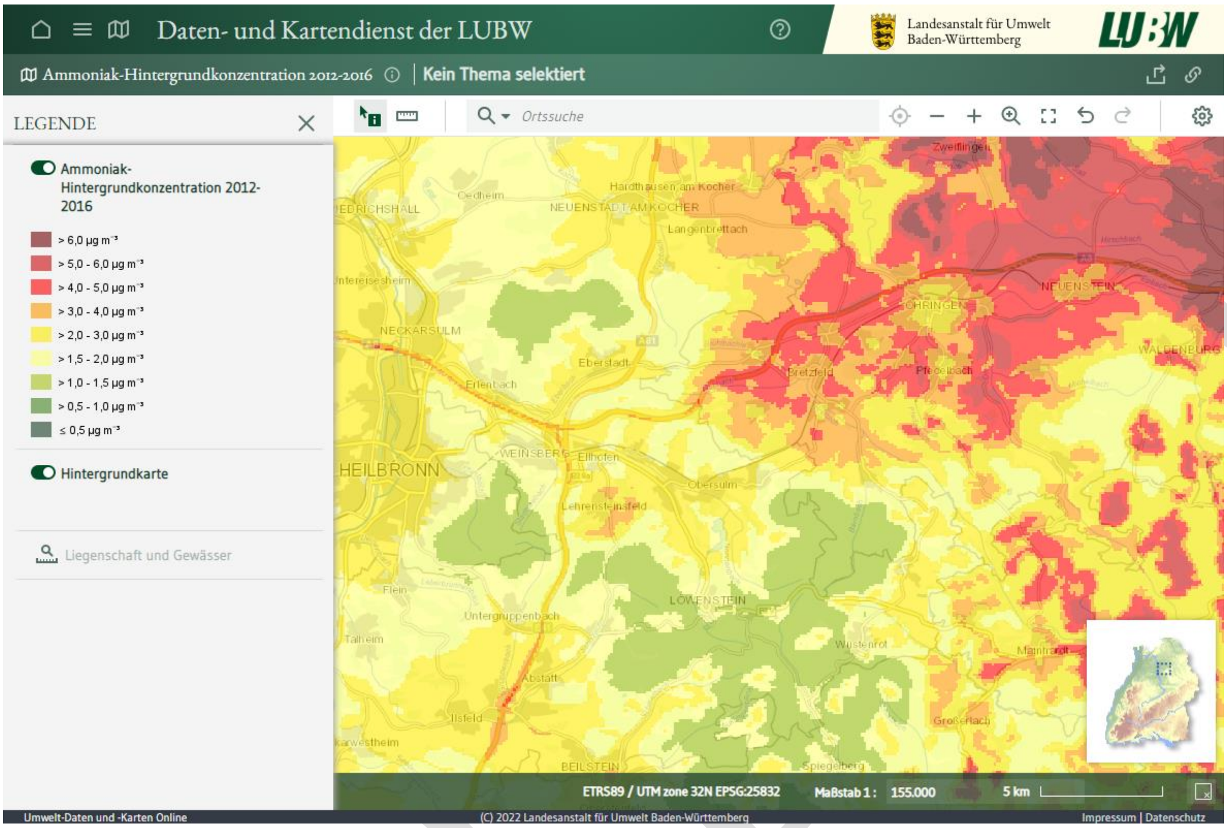
Introduction

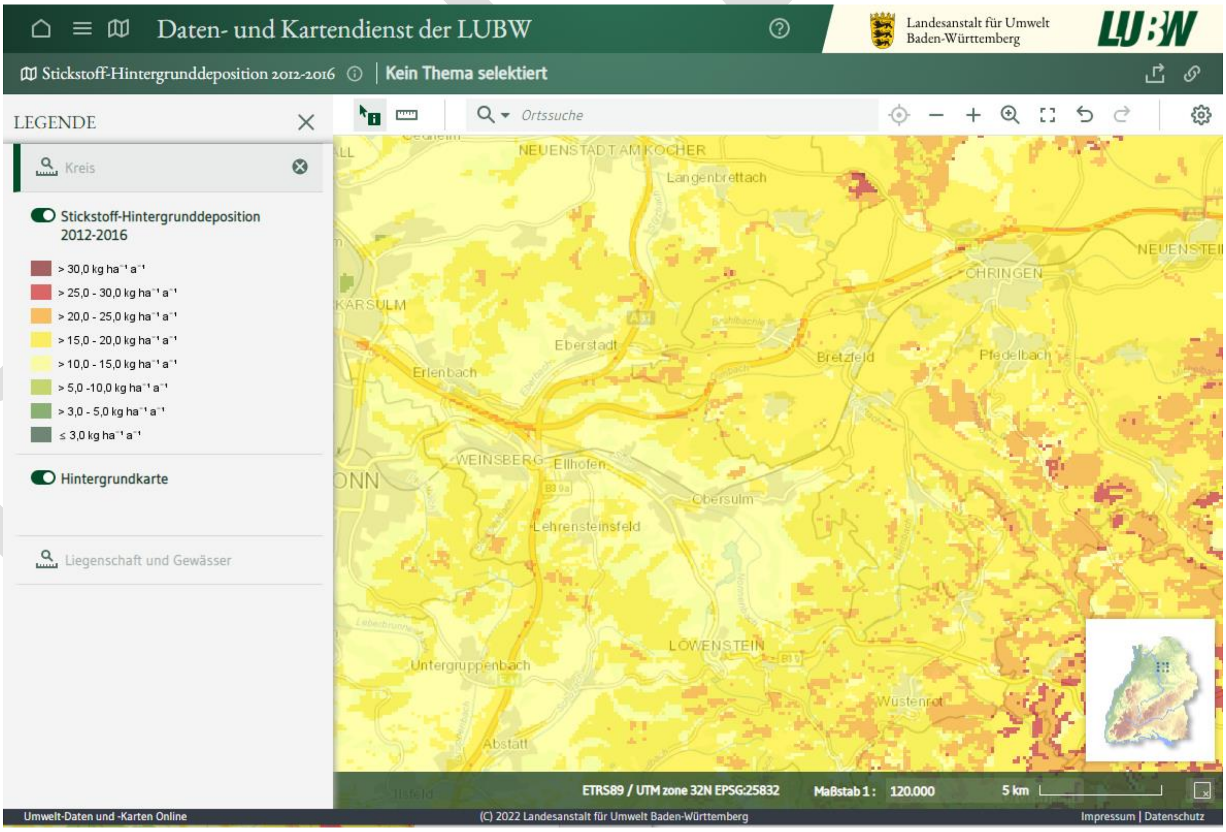
With this contribution, we suggest a straightforward approach to consider atmospheric ammonia concentrations and nitrogen deposition in the implementation of the Habitats Directive. According to the Natura 2000 Habitats Directive 92/43/EEC, the German federal states must take measures to achieve or maintain the favourable conservation status of habitat types listed in Annex I. Critical levels for ammonia and critical loads for nitrogen deposition are used to assess whether there is a need for the reduction of nitrogen inputs or whether additional input is still possible. Based on the research program StickstoffBW (2019), a habitat trophic standard for critical levels and critical loads is being developed. First estimates of habitat type critical levels are published in the national habitat handbook (BfN 2021, 2022). There are three reasons to use critical ammonia levels as a leading parameter in addition to the nitrogen deposition to implement Directive 92/43/EEC: 1. Habitat trophy management (up to 10 km from agricultural sources), 2. Indicator of the nitrogen saturation of vegetation, 3. direct effect on cryptogams (lichens and mosses) and indirect effect on ferns, flowering plants, insects and mycorrhiza. In Baden-Württemberg (BW), maps of the ammonia concentration and nitrogen deposition with a resolution of 100 m x 100 m are used as the data basis to assess the current pollution (Figure 24).

We propose "trophic levels" for the application of critical levels and critical loads according the following definition. A trophic level (TR) is the long-term target level below that a favourable conservation status concerning air-borne and soil-borne nitrogen is possible. TR's are defined for all habitat types. TR1 shows a low nitrogen supply and is typically characterized by the occurrence of highly nitrogen-sensitive plant and animal species. TR3 shows a comparatively high nitrogen supply and highly nitrogen-sensitive species are absent, but the general species composition still corresponds to a defined habitat type according to Directive 92/43/EEC. TR2 shows an intermediate nitrogen supply. The cause for high nitrogen supply can be natural (e.g. inherently high soil fertility, strong biological nitrogen fixation) or anthropogenic (e.g. former fertilization, high deposition or lack of biomass export). When setting the trophic level parameters, we assume that growth will not be limited by other factors. Growth conditions and hence biomass production typically increase from TR1 to TR3.

There is a strong interrelation between critical levels and critical loads: 32% - 49% of the nitrogen deposition in BW is caused by ammonia dry deposition (10th - 90th percentile, data from 44 BW districts; StickstoffBW 2022).

Figure 24: Ammonia concentration and nitrogen deposition in Baden-Württemberg, average for the years 2012-2016





Source: LUBW (2020).

This implies that a critical load of 20 kg N ha yr^{-1} corresponds to a NH_3 critical level of 2.0 – 3.1 µg m^{-3} (10th - 90th percentile), when a deposition velocity v_d of 1.2 cm s^{-1} (typical for lowland hay meadows in BW, habitat 6510) is assumed.

We will derive the habitat-specific critical level of NH_3 from the habitat-specific critical loads for nitrogen considering the trophic level with the help of the following criteria: 1.0 - 2.0 µg m^{-3} if a high proportion of very sensitive cryptogams, insects or mycorrhiza belong to the characteristic habitat species and 2.0 - 3.0 µg m^{-3} if less sensitive cryptogams, insects or mycorrhiza belong to the characteristic habitat species. 1 µg m^{-3} is taken as the field detection limit (minimum) and 3 µg m^{-3} is taken as the upper limit to protect vegetation as a nitrogen sink (maximum), also taking into account a maximum critical load of 20 kg ha yr^{-1} and a v_d of 1.2 cm s^{-1}.

Outlook and conclusions

The future assumptions, formulas and rules for the habitat-specific critical levels and critical loads are currently being compiled in a technical guidance document (update of StickstoffBW 2019). In future, specific attention should be paid to the atmospheric ammonia concentration as an easily measured key parameter for habitat trophy management (considering critical loads). Biomass thresholds for the different trophic levels and removal of N by biomass need to be assessed more accurately. Biomass production is the most important trophic indicator. Long-term integrated field monitoring of sensitive species including cryptogams, mycorrhizae and insects (not just ferns and flowering plants) with respect to trophic levels is needed for all sensitive habitat types. Establishing a general habitat trophic standard based on critical levels and critical loads is required for the implementation of directive 92/43/EEC. Critical surplus values for agricultural land should follow soon. We consider our suggested approach as a basis for further discussion among and within research institutions, consultant offices and federal and state administrations.

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4.2.7 Ammonia concentrations in Switzerland compared to critical levels and loads

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Measurement network

In 2020, ammonia passive sampler measurements in Switzerland and the Principality of Liechtenstein included at total of 89 sites. 13 sites have been operational since 2000, eight were added in 2004, 11 in 2008, 31 in 2013 and the remaining afterwards (Seitler 2021a; Seitler & Meier 2022). While many sites are situated in rural areas characterized by intensive livestock and crop farming, the measurement network also covers extensive alpine pastures, bogs and fens as well as traffic and urban sites. Additional measurements of ammonia and other nitrogen compounds are part of the National Air Pollution Monitoring Network (NABEL 2020). They include continuous NH_3 measurements at three EMEP sites (CRDS), daily concentration of the sum of reduced (NH_3 & NH_4^+) as well the sum of oxidized (HNO_3 NO_3^-) nitrogen compounds at two EMEP sites (impregnated filters), 2-weekly concentrations of NH_3, NH_4^+, HNO_3, NO_3^- at four sites (three EMEP sites, minidenuders) and wet deposited NH_4^+ and NO_3^- at three sites (two EMEP sites). Further measurements are carried out by cantons (bulk deposition (Seitler 2021b), minidenuders).

Ammonia concentrations

The highest annual ammonia concentrations are found in areas with intensive animal production (6–10 µg/m³) followed by areas with predominant crop farming (2–5 µg/m³). Peak concentrations occur during periods of slurry application. Measurement sites within a few kilometres may show different concentration values, but nevertheless show a parallel pattern over time. Besides a relatively large year-to-year variability, influenced by meteorological conditions, measurements show no change in average ammonia concentrations in the last 20 years. Preliminary analysis including data on NH_4^+ indicates, however, a slight decrease of total reduced nitrogen compounds.

Critical Load and Critical Level exceedances

On average in Switzerland, about half of the NH_3 concentration at a sensitive ecosystem is caused by sources within a radius of 4 kilometres; the other half is due to regional background (EKL 2014). In rural areas with intensive agriculture, ammonia dry deposition accounts for the major part of total nitrogen deposition. In other regions, total nitrogen deposition is largely influenced by wet deposition of NH_4^+ and NO_3^-. As a consequence, Critical Loads for nitrogen are more often exceeded than Critical Levels for ammonia (Rihm and Künzle 2019, BAFU 2020). The Federal Commission for Air Hygiene therefore concluded that the need for action to protect sensitive ecosystems becomes much more apparent in an assessment based on total nitrogen deposition than in an assessment based on gaseous ammonia concentrations only.

Ammonia concentration and lichens

A Swiss study (Urech et al. 2015) analysing the relationship between lichen vegetation and nitrogen pollution found that occurrence of nitrogen sensitive species steeply decreases above ammonia concentrations of 1 µg/m³. Based on lichen data over a period from 2001 to 2010 from 1815 trees in 10 different regions covering an ammonia gradient from 1 to 9 µg/m³ the authors concluded that the Critical Level for lichens needs to be substantially lower than 3 µg/m³.

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4.3 Workshop Discussion

4.3.1 Discussion points

During the workshop and the final discussion, it became clear that the disussing workshop participants see ammonia critical levels as a very import measure for ammonia mapping and ecosystem protection. It was worked out that critical levels play an evenly important role as critical loads and that both approaches are instruments that need to be differentiated and looked at. Besides the critical levels for ammonia levels for NO_x might also need revision.

The workshop participants agreed that in many countries there is a lack with regard to ammonia monitoring and urge that ammonia is considered in the future review and revision of the EU Air Quality Directive. Furthermore, a Europe-wide standardization of ammonia measurements would be a useful addition to verify the effectiveness of mitigation measures needed to meet the reduction commitments of the NEC Directive.

With regard to the questions raised prior to the ammonia workshop, some answers could be collected:

- **Is the use of passive samplers suitable? Are there devices to continously monitor NH_3 in the low range?**

The workshop participants agreed that the easy and cheap use of passive samplers is a well-established method for NH_3 measuring. It was stated that the measuring height plays an important role and that measuring should therefore always take place at a defined and standardized height (compare with European standard). Measuring ammonia in the low concentration range is generally seen as difficult, in part due to the stickiness of ammonia to instrument surfaces of real time monitors. It is recommended to monitor ammonia additionally with the denuder method or continous online analysers in order to calibrate the passive sampler employed in the field.

It is worth noting that as a consequence of the changing partitioning between gaseous NH_3 and aerosol NH_4^+ the gaseous NH_3 trends do not necessarily reflect the changes in ammonia emissions. Additional monitoring of NH_4^+ trends (aerosol and precipitation) is important for making such a comparison (cf. Key Finding 5, chapter 4.3.2).

- **Do you perform or are you aware of research on ammonia effects using controlled fumigation?**
- **Do you perform ammonia fumigation studies in the field?**

New research using controlled fumigation is planned in Sri Lanka by UK CEH. The study site for the set-up of a fumigation system is a woodland and the imapct on the site-specific vegetation shall be investigated. Furthermore, in 2021, CEH has established a new field ammonia fumigation system at Glencorse Field Facility (Southwest of Edinburgh). This site releases ammonia at a similar level to Whim, but assesses the impact of ammonia within a woodland environment. The concentration range goes up to 40 $\mu g\ m^{-3}$.

- **Do you perform or are you aware of gradient studies using active or passive biomonitoring?**

Gradient studies mainly using passive biomonitoring with lichens are planned in Finland (S. Manninen) and in the UK (M. Jones) using e.g. Alpha samplers or CRDS analysers. In this context, the question was raised how to differentiate between oxidised/reduced N compounds adding to N deposition, i.e. how to evaluate the sole effect of ammonia on vegetation and species composition in field studies.

- **Would you support annual, monthly and/or short-term CLe?**

It was suggested that the provisional monthly CLe of 23 was too high, but new findings that support or contradict the value are missing. Generally, CLe for annual ammonia are supported due to missing. sufficient new evident to change them.

- **Would you support to include knowledge on CLo into scientific justification of CLe?**

It was recognized that there is a relationship between critical loads and levels that can be used in the assessment of NH_3 effects on vegetation and species composition change. For the conversion, assumptions on the prevailing NH_3 deposition velocity and on the ratio of NH_3 deposition to other non-NH_3 N deposition fluxes at a location need to be made (e.g. taken from model results). It was shown that critical levels from critical loads are in the same range as existing CLe between 1-3 µg and therefore give further support to the robustness of the existing CLe. All participants found it important to keep both approaches, but there need to be further discussions on whether or not to include information on the relationship between the two metrics into the Mapping Manual.

- **Is there enough evidence for changing/keeping the CLe?**

As only few new studies address the effects of ammonia concentrations on plants, all participants agreed that more studies, including higher and lower species are needed. Field fumigation studies should cover different climatic conditions and ecosystem types and the established vegetation as well as biomonitoring plants should be involved. Particular attention should be paid to important habitat-specific species.

However, based on the few new findings, the annual critical levels of 1 µg m^{-3} for lichens and bryophytes and of 3 µg m^{-3} for higher plants can be confirmed, i.e. there is no evidence to change these values. In this context, it was also stated that the long-term ammonia CLe are of higher relevance than the monthly CLe that were referred to as provisional values in the 2006 Mapping Manual. Regarding the monthly CLe of 23 µg m^{-3} there are no new findings that support or contradict these values.

- **Which further steps after the Workshop should be taken in the review of ammonia CLe?**

Workshop recommendations and conclusions, in particular for the Air Quality Directive (see key findings, chapter 4.3.2)

Further information/reading:

- EMEP data to be included (consider data exclusion rules such as data has to be gathered for at least 75% of the year, at least one month per season) is very welcome. Please contact Wenche Aas or Kjetil Tørseth at CCC/NILU for more details
- ICP Vegetation review on NO_2 Critical Levels (first online meeting 24th May 2022) → contact Mike Perring if interested
- Standard method for the determination of the concentration of ammonia using diffusive samplers: https://www.en-standard.eu/csn-en-17346-ambient-air-standard-method-for-the-determination-of-the-concentration-of-ammonia-using-diffusive-samplers/
- Ammonia comic in 16 languages: https://www.ucd.ie/ammonian2k/publications/comic/
- Papers on Gradko measurements:
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  https://www.sciencedirect.com/science/article/pii/S1352231020302624
- NI-wide network data will be published in https://eidc.ac.uk/
  This is the Environmental Information Data Centre (EIDC) is part of the Natural Environment Research Council's (NERC) Environmental Data Service and is hosted by the UK Centre for Ecology & Hydrology (UKCEH). EIDC manages nationally-important datasets concerned with the terrestrial and freshwater sciences
- More information on Ireland's NEC network design:
  https://researchrepository.ucd.ie/bitstream/10197/12330/3/National%20Ecosystem%20Monitoring%20Network%20%28NEMN%29-Design.pdf
- Paper on different passive samplers:
  https://www.sciencedirect.com/science/article/abs/pii/S1352231018308185?via%3Dihub#fig4
- Link to report on decision making thresholds:
  https://hub.jncc.gov.uk/assets/6cce4f2e-e481-4ec2-b369-2b4026c88447
- Research in ecotrons in V-Connecterra (University of Belgium) in Natura2000 areas:
  https://www.terhills-nationaalparkhogekempen.be/over-connecterra/veldstudiecentrum/608

4.3.2 Key findings

The following **key messages** and **recommendations** emerged from the Dessau Ammonia Workshop:

1. The workshop participants agreed that ammonia should be better respected in the future environmental legislation. At present, there are no ammonia air quality limit values set in the European Air Quality or Habitat Directive. Given the upcoming review of the AAQD, and recognizing the inclusion of impacts on ecosystems as an essential part of both directives, it is recommended that future revisions of the AAQD directives should contain a Europe-wide standardization of ammonia measurements, to verify the effectiveness of mitigation measures needed to meet the reduction commitments. A consistent monitoring of ammonia with harmonization of data gathering and processing strategies across Europe will help to improve EU policies by laying the basis for further understanding and assessment of the effects on ecosystems, on plants and plant communities.

2. The monitoring of air pollution impacts under Article 9 (and Annex V) of the National Emissions reductions commitments Directive (NECD) offers the possibility to include NH_3 monitoring data. In contrast to several soil and vegetation parameters ammonia critical levels or air concentration measurements are not explicitly mentioned. The workshop participants concluded that the reporting of ammonia concentrations in relation to the required vegetation and soil data could be a good complement with regard to the observation of the effectiveness of the directive and that a standardized protocol should be developed in the near future.

3. Air quality limit values for human health include total $PM_{2.5}$ and there are substantial nitrogen-related contributions to $PM_{2.5}$ through a) reaction of NO_x emissions, b) reaction of NH_3 emissions and c) ammonia-stimulated oxidation of SO_2 emissions to SO_4^{2-} levels. The successful reductions of SO_2 and NO_x emissions slow down the formation rate of secondary inorganic PM, which is reflected in substantially reducing trends of ammonium (NH_4^+) in $PM_{2.5}$ in Europe and North America over the last 20 years. At the same time, ammonia residence times are increased, which is reflected in the slight upward trend in levels seen on both sides of the Atlantic.

4. European standards for passive monitoring of ammonia have been published by the Comité Européen de Normalisation (EN 17346 (2020)), and metrological comparisons of widely used passive ammonia samplers have been performed within the European MetNH3 project. The workshop participants agree that a wide range of passive sampler implementations may achieve cost-effective monitoring of ammonia with typically monthly and bi-weekly time-integrated sampling, for the purpose of assessing long-term trends and spatial patterns. However, the performance of a method depends on specific laboratory implementation. For the setup of a monitoring network, therefore it was recommended to include parallel sampling with a recognized monitoring device on a continuous basis across the range of concentrations at selected sites, to allow ongoing calibration of passive samplers against a reference method. Currently, various real-time monitors for low concentrations of ammonia are being introduced to the market, which can be used for the calibration of passive samplers.

5. The area-wide exceeding of the compensation point due to high fertilization of croplands and grasslands and substantial emission-reductions of SO_2 and NO_x have altered the partitioning between gaseous NH_3 and aerosol NH_4^+. For this reason, the assessment of trends in ammonia emissions and concentrations requires simultaneous measurement of the nitrogen surplus in

agriculture, gaseous NH_3, particulate matter NH_4^+ and of NH_4^+ in rain. Several datasets show gaseous NH_3 increasing while $PM_{2.5}$ NH_4^+ is simultaneously decreasing (s. above). Higher levels of ammonia may also be due to stronger volatilization caused by rising temperatures. The workshop participants therefore strongly encourage to continue long-term studies on changes in the chemical and physical climate and their relationships to the biosphere. Future research should also make use of remote sensing technologies. The initiative of the European Space Agency to develop the NitroSat candidate mission for 2030 is greatly to be welcomed, and if finally selected, would provide unprecedented improvement in understanding the fine scale distribution and trends of ammonia and other nitrogen gases globally.

6. There have only been few long-term studies on the effects of elevated NH_3 concentrations on vegetation in the last 15 years. Existing studies covered in an extensive review (see chapter 2 for documentation) broadly support the ammonia critical levels established by the 2006 UNECE Ammonia Workshop. However, it is noted that further long-term experimental studies at a field scale for different habitat types and climates must be a priority. Under a changing pollution climate (with less SO_2 and a more alkaline atmosphere and climatic change), there is a risk that some of the most sensitive species groups (e.g. higher species, certain lichens, mycorrhiza or insects) may become more sensitive to high NH_3 concentrations. The workshop participants therefore advocate for efforts to include ammonia and nitrogen deposition in existing ecological research networks, e.g. the LTER sites. Furthermore, more basic research is needed e.g. in the NH_3 driven distortion of algae and fungi in lichens and the metabolism of ammonia/ammonium in higher plants.

7. Reduced and oxidized nitrogen forms and their adverse ecological and environmental effects cannot be disentangled from each other. While the last revision of ammonia took place in 2006, the UNECE Critical Level for NO_2 has not been reviewed for 25 years and it is unlikely to be sufficiently precautionary. Since it was originally set for "average" plant species and crops, it may not be suited to protect the most sensitive species, e.g. lichens and oligotrophic higher species. A process to review the UNECE NO_2 critical level has recently been started and will yield important information for the reduced nitrogen species.

8. The workshop confirmed the validity of annual UNECE Critical Levels (CLe) of 1 $\mu g\ m^{-3}$ for lichens and bryophytes and of 3 $\mu g\ m^{-3}$ for higher plants. CLe account for both direct (e.g. toxic) and indirect effects (e.g. species composition). Although species composition change is more associated with N-deposition and eutrophication, the mathematical conversion of critical nitrogen deposition values to NH_3 concentration with standardized meteorological assumptions showed a good agreement with the UNECE Critical Levels, giving further evidence for their scientific robustness.

9. Specific points should be clarified with respect to the UNECE ammonia critical levels agreed in Edinburgh in 2006, which focus on long term protection of ecosystems and sensitive organisms:

a) The 'long-term' critical levels, expressed as annual mean NH_3 concentrations of 1 and 3 [2-4] $\mu g\ m^{-3}$, cannot be assumed to give protection for long periods of more than 20 to 30 years.

b) The monthly critical level of 23 $\mu g\ m^{-3}$ had been derived for higher plants, and does not apply to lichens, bryophytes and ecosystems where these are important to ecosystem integrity, e.g. peatlands. It can easily be seen that one month of 23 $\mu g\ m^{-3}$, would give a minimum annual average of 1.9 $\mu g\ m^{-3}$, which already exceeds the long-term critical level. To be mathematically consistent with the long-term critical level, a maximum monthly value of 12 $\mu g\ m^{-3}$ would apply, though further evidence would be needed to assess whether this value is sufficiently precautionary for sensitive lower and higher plant species.

10. To deal with the above-mentioned uncertainties, the workshop participants outlined the need for further basic and field-based research to refine critical levels. While ammonia field fumigation systems have been established in peatlands and forests in the UK, research infrastructures are not available in other countries and in other nitrogen-limited ecosystems, e.g. highly protected acidic and calcareous grasslands. Besides the set-up and technical improvement of ammonia fumigation systems in established vegetation, studies in the lee of livestock facilities using plants (on-site and exposed) along the ammonia gradient will create new dose-response relationships that may be used to update critical levels in the future. Studies should focus on single species as well as on species interactions, which will in the long-term lead to the disappearance of the sensitive components at the expense of the nitrogen loving species.

Meanwhile, on 4th of April 2022, these workshop recommendations have been presented by UBA at the EU Stakeholder consultations workshop on the revision of the AAQD (Policy area 3: Strengthening air quality monitoring, modelling and plans).

The note to the EU Stakeholder Consultations was as follows.

The Ammonia Expert Workshop 2022, held in the framework of the Convention on Long-Range Transboundary Air Pollution on 28 and 29 March 2022 (Dessau/online) draws the following conclusion and recommendations in relation to the revision of the EU Ambient Air Quality Directives:

Ammonia impacts ecosystems and contributes to the loss of biodiversity. In addition, it is a key precursor in the formation of PM2.5. It is likely that critical levels of ammonia in the ground-level air layer, as set by Convention on Long-Range Transboundary Air Pollution (CLTRAP) and as confirmed by the review of recent data within the above mentioned 2022-workshop, are exceeded for a substantial number of ecosystems across Europe.

The workshop points out the deficiency that at present there is no obligation on ammonia monitoring and procedure for assessment set in the Ambient Air Quality Directives. Given the ongoing process to revise the AAQDs, and recognizing that impacts on ecosystems and vegetation are already now an essential part of the directive (as already reflected in ozone, NOx, and SO_2 assessments), the workshop members urge that ammonia is considered in the future review and revision of the AAQD.

Addressing ammonia in the AAQDs would harmonize EU legislation with the NEC Directive and is essential for effective implementation of the Habitats Directive. A Europe-wide standardization of ammonia measurements, would be a useful addition to verify the effectiveness of mitigation measures needed to meet the reduction commitments of the NEC Directive. Furthermore, a consistent monitoring of ammonia with harmonization of data gathering and processing strategies across Europe will help to improve EU policies in three ways:

(1) It will help to lay the basis for further understanding and assessment of the effects on ecosystems, on plants and plant communities.

(2) Available data will help to improve chemical-transport models through evaluation and validation.

(3) and lastly the combination of both (1) and (2) will lay the basis for effective reduction strategies.

A low-cost method for the determination of the concentration of ammonia using diffusive samplers is given by the European Standard method EN 17346.

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