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Directional passive ambient air monitoring of ammonia for fugitive source attribution; a field trial with wind tunnel characteristics

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HIGHLIGHTS

- A Directional Passive Air Sampler (DPAS) is proposed for ambient ammonia monitoring.
- The DPAS was tested in the field with a wind-dependent artificial release of ammonia.
- The sampling of the DPAS-MANDE system showed distinct directional discrimination.
- The MANDEs within the DPAS are subject to diffusional interference and can saturate.
- The DPAS-MANDE system may be used to identify and target fugitive sources of ammonia.

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ABSTRACT

Atmospheric ammonia is a precursor for secondary particulate matter formation, which harms human health and contributes to acidification and eutrophication. Under the 2012 Gothenburg Protocol, 2005 emissions must be cut by 6% by 2020. In the UK, 83% of total emissions originate from agricultural practices such as fertilizer use and rearing of livestock, with emissions that are spatially extensive and variable in nature. Such fugitive emissions make resolving and tracking of individual site performance challenging. The Directional Passive Air quality Sampler (DPAS) was trialled at Whim Bog, an experimental site with a wind-controlled artificial release of ammonia, in combination with CEH-developed ammonia samplers. Whilst saturation issues were identified, two DPAS-MANDE (Mini Annular Denuder) systems, when deployed in parallel, displayed an average relative deviation of 15% (2–54%) across all 12 directions, with the directions exposed to the ammonia source showing ~5% difference. The DPAS-MANDE has shown great potential for directional discrimination and can contribute to the understanding and management of fugitive ammonia sources from intensive agriculture sites.
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1. Introduction

Ambient air monitoring of ammonia is a priority issue due to its potentially harmful effects on human health, as it is a precursor for secondary particulate matter formation; and ecosystems, as it contributes to acidification and eutrophication (Behera et al., 2013; Bessagnet et al., 2014; Cape et al., 2009; Phoenix et al., 2012; Sheppard et al., 2011). Because of its potential for chemical reaction in the atmosphere, ammonia not only has a localised impact, but it contributes to the formation of other compounds, which can contribute to long-distance effects. International policy drivers to improve the environment include the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP Gothenburg Protocol) and the EC National Emission Ceilings Directive 2001/81/EC (NECD). Under the 2012 revision of the Gothenburg Protocol, EU member states must jointly cut their emissions of NH_3 from base 2005 to 2020 by 6%.

After 1950, the introduction of the Haber-Bosch process (for artificial nitrogen fixation), increased emissions from fertilizer use and intensive livestock production systems (Erisman et al., 2008). Today, the main emissions into the atmosphere in the UK originate from agricultural practices, 83% in 2014 (NAEI, 2014).

Ambient concentrations of ammonia vary as a function of climate, season and local sources. “Background” concentrations, i.e.

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those due to mostly biogenic emissions, are low, in remote regions <0.5 μg m^{-3}, whereas in urban and agricultural areas average concentrations are generally significantly higher than this and in intensive regions can be > 10 μg m^{-3}. Close to point or area sources of ammonia, concentrations can be much higher (Loubet et al., 2009; Staebler et al., 2009; Vogt et al., 2013; von Bobrutzki et al., 2012; von Bobrutzki et al., 2010).

Ambient monitoring is required for management and control. Ammonia monitoring over longer periods is often done using air samplers to capture ammonia with off-line analysis of the ammonia as ammonium (NH_4^+). Samplers include filter packs, denuders and passive samplers. Passive samplers collect ammonia by diffusion, and typically provide a time resolution of one week to three months. Active samplers are attached to a pump ensuring a continuous air flow and temporal resolutions of the order of 1 h−1 month are achievable. Higher time resolution on-line measurement techniques are available but many are still research items rather than routine monitoring tools. Instruments include denuders coupled to wet chemical analyzers, infrared and UV spectroscopy, chemiluminescence and chemical ionization mass spectrometry (CIMS). Active and high-temporal resolution monitoring devices tend to be bulky, costly and require power and calibration; however they can provide source identification combined with local activity knowledge and meteorology. Passive samplers tend to be cheaper and easier to locate, but provide lower temporal resolution and are generally non-directional.

Ammonia sources are mainly fugitive, with spatially extensive and variable source processes, making monitoring and attribution challenging unless directional monitoring and analysis is carried out. In complex industrial environments, regulators and operators need simple techniques for resolving and tracking the emissions performance of individual sites, so that limits can be met and improvements can be targeted appropriately (Ferranti et al., 2014).

Directional passive air quality samplers (DPASs) are proposed here as complementary monitoring tools that can contribute to the understanding and management of fugitive ammonia sources from intensive agriculture sites. The DPAS is an unpowered monitor that pivots in the wind to collect directional samples for analysis and attribution. DPASs are flexible to deploy, not bulky, do not require power, and can be used in arrays to determine the incremental impacts of pollutants from specific activities, e.g. around a source.

The DPAS performance was assessed at Whim Bog, Edinburgh. This experimental site is an ombrotrophic bog site which is used to compare the effects of dry and wet nitrogen deposition on vegetation (Leith et al., 2004; Sheppard et al., 2011). The site has an artificial line source of ammonia gas, which is coupled to wind direction and wind speed and logged. The DPAS was tested here because there is one known and well characterised ammonia source and meteorology is measured continuously. Two types of ammonia samplers were tested inside the DPAS, both of which have been developed by CEH Edinburgh: the CEH Adapted Low-cost Passive High Absorption (ALPHA) sampler (Tang et al., 2001) and the CEH Mini Annular Denuder (MANDE) flux samplers (Tang, pers.comm.).

DPAS testing was carried out from Nov 2012–Apr 2013 with five exposure periods. This paper summarises the results from these experiments, which were carried out to assess the potential for the DPAS-ALPHA and the DPAS-MANDE systems to collect fugitive ammonia with directional discrimination.

2. Methodology

2.1. Directional passive air quality sampler (DPAS)

DPAS (Patent Number US 8,413,527 B2) is the result of collaboration between Lancaster University and the Environment Agency for England. It has been developed for environmental applications over the past seven years, which have involved different pollutants of interest, namely NO_2 (Lin et al., 2011a, 2010a, 2010b) and particulates (Ferranti et al., 2014; Lin et al., 2011b).

The current prototype (4, Fig. 1) has improved the previous design (Ferranti et al., 2014), by manufacturing it with a stronger material, improving pollutant collection with a wider mouth, reducing internal gaps to decrease air circulation in sheltered channels, and making it more wind responsive with a larger fin and lower friction bearings. As a result of these improvements, internal wind speeds are now 92% of external wind speeds (88% for prototype 3) and DPASs now align with the wind for windspeeds down to ~1.5 m/s (~2.2 m/s for prototype 3).

The DPAS consists of rotating and non-rotating parts (Fig. 1). The fixed sampling carousel has 12 channels that collect samples to discriminate pollutant fluxes from 12 × 30° directions. The centre of the carousel was designed to be used as location for blanks; in particular, it was designed to fit 3 vials for particulate sampling. The DPAS is mounted on a stand, and is aligned to North. DPAS samplers are levelled using a flat slab base, weighed down with concrete blocks (Fig. 2).

The 'flux sampler that turns to face the wind' approach has been used before (Leuning et al., 1985), although sampling omnidirectionally instead (only one sample is collected). This earlier study focused on ammonia sampling using an oxalic acid coating over a much larger surface area based on a longer and spiral-wound geometry. The DPAS is more versatile in that it pivots in the wind to collect 12 directional (30°) samples, which are analysed separately. Also it can be used to collect other pollutants too (as described earlier), by changing the sampling media.

2.2. Ammonia passive samplers and analysis

Two types of ammonia samplers were used inside the DPAS (Fig. 3), ALPHAs and MANDEs. The CEH ALPHA sampler was developed by the UK Centre for Ecology and Hydrology (Tang et al., 2001) and has been used in the UK National Ammonia Monitoring Network (NAMN) since 2000. Conventional sampling consists of three replicate ALPHA samplers mounted with velcro to an aerodynamically shaped support (upturned frisbee or plant saucer) on a pole or post at a sampling height of about 1.5 m above ground level. Anti-bird spikes are mounted on top to deter birds from perching. Triplicate ALPHAs are used to provide an assessment of the precision of the ammonia air concentration measurement.

MANDEs have also been developed at CEH, based on the design by Possanzini et al., 1983 Each MANDE is 5 cm in length and consists of coaxial borosilicate glass tubes: a smaller tube (3 mm id x 5 mm od) suspended inside a larger tube (7 mm id x 10 mm od) coated on the inside with citric acid. The configuration forces air stream to flow through the annular space between the outer and inner tubes, increasing the surface area for retention of gaseous NH_3. Each MANDE is connected in series to a 2.3 cm long glass inlet with a thin stainless steel disc having a 1 mm orifice in the centre (Schjoerring, 1995). This orifice is expected to decrease air flow speed inside the MANDE, to achieve low friction velocity and provide high NH_3 collection efficiency. The MANDE, with the 1 mm orifice at the inlet, is deployed as a flux sampler inside the DPAS, following the principles of passive flux sampling described by Schjoerring (1995).

The Leuning et al., 1985 sampler also used an orifice to reduce the flow through the sampler, based on the same principle as the MANDE, and was proven to collect 100% of the ammonia that entered the sampler for a wide range of ammonia fluxes, so that the air leaving the sampler was effectively stripped of its ammonia

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