Monitoring Ambient Air 2005:
Diffusive Monitoring, Techniques and Applications
Conference with Exhibition

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MEETING THE AMBIENT AIR DIRECTIVES

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ABSTRACT

The presentation will first give an outline of the Clean Air for Europe (CAFE) process, initiated by the Community’s 6th Environmental Action plan. The Commission adopted its main deliverable, the Thematic Strategy on Air Pollution, on 21 September 2005. The strategy is defining the intermediate air quality objectives for the Community until 2020, based on the evaluation of potential for air pollution abatement, its costs and the benefits in terms of public health and ecosystems improvement. Particulate matter and ozone are the principal target pollutants. Though current legislation is already delivering a lot of benefits and will continue to do so in the near future, it is estimated that in the year 2020 roughly 270,000 EU citizens will still prematurely die due to fine particles, if nothing further is done.

An important measure outlined in the strategy, which is already on its path through the Council and the Parliament, is the revision of the air quality directives. Informing on the revision will form the core of the presentation.

Air Quality Framework Directive, 1-3 Daughter Directives and the Exchange of Information Decision are in the proposal all merged into one directive. The presentation will try to present the rationale for the proposal and some of its provisions, in particular from the perspective of the current experience with the directives. It will for example briefly address the difficulties with meeting the PM10 limit values.

The proposal retains all existing monitoring provisions as well as the limit values. The indicative PM10 limit value is replaced by a proposal for a PM2.5 concentration cap of 25 µg/m³ and a target of 10-year 20% reduction of exposure to fine particles in the Member State, as assessed by the concentrations at urban background locations. Proposal prescribes specific PM2.5 monitoring provisions as well as provisions on how to account for natural contribution and/or apply for possible time extension on the attainability of the limit value for up to five years, based on specific conditions. The Commission, assisted by the Data Exchange Group, is currently preparing the new reporting provisions. Changes will include a move towards a single monitoring dataflow and specific GIS based requirements for reporting on the assessment throughout the territory. It is anticipated that the new reporting provisions will represent a qualitative leap towards the implementation of INSPIRE in the air quality field.

In line with the main topic of the conference, a part of the presentation will be devoted to the role of diffusive monitoring within the existing directives as well as the new proposal.

The strategy, directive proposal and a number of accompanying reference documents, such as the WHO advice to CAFE, impact assessment and the cost-benefit analysis can be found at http://europa.eu.int/comm/environment/air/cafe/index.htm.
DIFFUSIVE SAMPLING
WHAT’S NEW?

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ABSTRACT

Diffusive sampling appears to have reached a state of maturity. A survey of recent literature sources reveals that publications in which diffusive sampling plays a key role focus mainly on applications, which are plentiful, e.g.:

- Assessment of pollutant levels in urban environments
- Assessment of personal exposure in the workplace, indoors and outdoors
- Assessment of spatial variability of pollutant concentrations, e.g. aimed at siting of monitoring stations
- Measurement of pollutant fluxes
- Establishment of indoor-outdoor relationships of pollutant concentrations.

New developments appear to focus on the search of optimal samplers for certain key pollutants: 1,3-butadiene, benzene, ozone, nitrogen dioxide, or on the search for new – although perhaps not diffusive - samplers (SPME and SPMD devices).

However, the developments in diffusive sampling have one thing in common: they are mainly results of individual efforts. As a consequence, little is known about the comparability of data obtained by diffusive sampling. This may be particularly problematic when these data are to be used for quantitative purposes, for cross-country comparisons, and not for relative purposes within single studies.

In this presentation information will be given about the state-of-the-art of diffusive sampling as a technique for quantitative evaluation of pollutant concentration levels and of further needs for developments and initiatives.
The concept of sampling ambient and workplace air for pollutants using the principle of diffusive sampling is well established. Diffusive sampling is featured in recognised validated monitoring procedures and is applicable for acid gases and VOCs. The principles utilised are based on the established gas laws of Boyle and Charles, and gaseous diffusion parameters developed by Graham and Maxwell. The paper will present a resume of the gas and diffusion laws and examine the application of Fick’s law to diffusive monitoring, practical monitoring devices and the influence of ambient factors such as humidity, temperature. The factors to consider in an overall monitoring exercise will be discussed including validation of methods and overall uncertainty in results. An outline of the instrumentation needed to measure pollutants (analytes) of interest will also be included. The application of diffusive sampling for monitoring SO₂ and NOₓ in ambient air will be used to illustrate the technique.
A CRITICAL APPRAISAL OF MONITORING TECHNIQUES

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ABSTRACT

About thirty years ago diffusive samplers were introduced to assess the air quality in the course of workplace surveillance in a convenient manner. The validity of workplace measurements by diffusive sampling had been shown in two EU conferences which took place in Luxembourg in 1986 and 1991.

Widening the scope of application the main areas of additional interest were ambient and indoor air quality. A good overview of this large field of application and the quality of data received was given at the "International Conference – Measuring Air Pollutants by Diffusive Sampling" in Montpellier in 2001.

This development was accompanied with the work of WG 11 of CEN/TC264 which compiled a series of European standards (EN 13528, 1 – 3, general and ambient air quality and EN 14412, indoor air quality) to set a reliable basis for validating and using diffusive samplers.

In parallel a variety of international laboratory and field tests were performed to prove the validity of different types of diffusive samplers to measure ambient air quality. Most of these tests were carried out on the basis of EN 13528 or/and in the course of the implementation of benzene references methods due to the EC Benzene Directive.

<table>
<thead>
<tr>
<th>Analyte</th>
<th>Organisation</th>
<th>Location</th>
<th>Date</th>
<th>Field-/Laboratory-Test</th>
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<tr>
<td>NH₃</td>
<td>GSF</td>
<td>Neuherberg, D</td>
<td>1998</td>
<td>Field</td>
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<tr>
<td>Benzene</td>
<td>NPL</td>
<td>Teddington, UK</td>
<td>2001</td>
<td>Laboratory</td>
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<tr>
<td>Benzene</td>
<td>VITO</td>
<td>Mol, B</td>
<td>2001</td>
<td>Laboratory/Field Condition</td>
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<tr>
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<td>VITO</td>
<td>Willebroek, B</td>
<td>2001</td>
<td>Field</td>
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<tr>
<td>O₃, NO₂, SO₂</td>
<td>NPL</td>
<td>Teddington, UK</td>
<td>2002</td>
<td>Field</td>
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<td>NMI</td>
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<td>2002</td>
<td>Field</td>
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<td>NO₂</td>
<td>ERLAP</td>
<td>Ispra, I; Paris, F</td>
<td>2003</td>
<td>Field + Laboratory</td>
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The appraisal of the validation campaigns due to the Benzene Directive comprehends the combined uncertainty for each method and different experimental conditions. Taking into account the mean value of all prevailing conditions, the combined uncertainties of diffusive and pumped sampling were considered to be equal with values of about 10%.

In addition to the reports of these large intercomparison exercises there are numerous publications and studies dealing with the validation of specific diffusive sampler types for different analytes and applications mainly on ambient and indoor air quality and personal exposure. For most of these applications diffusive samplers will be used as substitute of complicated, time consuming or expensive but well established methods. Generally, the usefulness and the validity of diffusive sampling results had been shown. On the other hand, individual drawbacks and advantages of the sampler types had been outlined, e.g. the dependence of the uptake rate on the velocity of adjacent air flows.
FORMATION AND FATE OF OZONE PRECURSORS

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ABSTRACT
Tropospheric ozone is of concern due to its noxious properties towards vegetation and humans. Levels of tropospheric ozone are dominated by photochemical formation from volatile organic compounds (VOC) and nitrogen monoxide and nitrogen dioxide (NOx). A series of VOC has been identified in Directive 2002/3/EC as ozone precursors reflecting mostly anthropogenic hydrocarbons. The term VOC covers a wide range of organic compounds, and the reactivity of VOC and their potential to form ozone varies substantially. Generally, the lifetime of biogenic VOC is much shorter than for typical anthropogenic VOC, and this affects atmospheric transport regimes (local to hemispheric scales) and fate of these compounds. On a global scale, natural emissions are the major source to VOC but the exact magnitude is highly uncertain. Thus it is important to assess the relative contribution from VOC of biogenic and anthropogenic origin to ozone formation, especially in relation to the above mentioned Directive.

Measurements of radiocarbon content of photochemical oxidation products can provide information about the sources. Such investigations have been performed for carboxylic acids and carbonyl compounds, and the results showed surprisingly high biogenic contributions (Glasius et al., 2001; Larsen et al., 2001).
Sources to ozone precursors have also been investigated using semi-continuous GC-analysis techniques and receptor modelling. In a recent study the ozone forming potential of C$_2$-C$_9$ hydrocarbons in Milan air was attributed equally to gasoline and diesel vehicle emissions (Latella et al., 2005).

The presentation will cover two areas:

Firstly, a brief history of national monitoring networks for hydrocarbons in the United Kingdom will be given. Large volumes of data were obtained over the period 1993 - 2000, when up to 13 sites generated hourly data for 25 hydrocarbon species very similar to the list of ozone precursors given in the Ozone Directive. Since then there has been a smaller automatic network with 3 sites monitoring BTEX and 1,3-butadiene, and 2 sites monitoring a larger range of hydrocarbons, and a larger network of 35 non-automatic sites generating fortnightly data for benzene only using a pumped method, with some sites also monitoring 1,3-butadiene using diffusive sampling. New calibration cylinder standards have been developed at NPL for automatic sites monitoring the full range of ozone precursors.

The second part of the talk will describe work at NPL to establish what information about ozone precursors can be reliably extracted from the pumped and diffusive chromatograms obtained from the non-automatic network. The results showed that of the 29 species in the Ozone Directive, data for 13 was available from the pumped samples, and data for 10 was available from the diffusive samples, with 2 species common to both methods. The results compare well with previous automatic data, and make an interesting comparison with emission inventories for the same species.
WHICH SORBENTS TO USE AND WHEN – A PRACTICAL GUIDE

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ABSTRACT

Sorbent selection: Correct selection of sorbents is critical to the success of any tube-based vapour monitoring exercise. Whether pumped (active) or diffusive (passive) sampling techniques are being employed, the two main objectives are invariably:

1. quantitative and complete retention of target analytes during sampling and .....  
2. complete release of target analytes during desorption & analysis  

A secondary objective may also be 

3. selective elimination of high concentration interferences (e.g. water) during sampling.  

Few people fundamentally understand all the processes involved in sorbent-sorbate interactions, so, for most of us, sorbent selection remains a happy mixture of science and art with a few guidelines/rules and known pitfalls to assist the otherwise empirical process of sorbent selection. Key parameters to consider when choosing sorbents are: - sorbent strength, reactivity (storage and desorption issues), artifact levels, hydrophobicity, temperature range, batch-to-batch variability, physical strength (tendency to form fines), desorption efficiency, known retention volume / uptake rate and impedance. A summary of the impact of each of these issues and respective guidance will be presented.

While diffusive sampling is conventionally restricted to one sampling surface (and thus a single sorbent), up to 4 sorbents can be successfully arranged in order of increasing sorbent strength for pumped monitoring. Such samplers are very versatile with respect to analyte volatility and are extensively used, however, use of multiple sorbents brings additional considerations – particularly issues of migration during storage, mis-matching of sorbents with different maximum temperatures and water retention. These issues and associated practical guidelines will also be discussed.

Validation of sorbent selection and sampler performance: Various methodologies have been described in the literature regarding validation of sampling efficiency. Examples include use of back up tubes (pumped monitoring), distributed volume sampling (pumped monitoring) and parallel monitoring of pumped with diffusive samplers & vice versa. However, validation of desorption efficiency during analysis is equally important. This used to be a difficult and imprecise art relying on generating comparative data between liquid injection and thermal desorption under identical analytical conditions. This is one of the areas where recent developments in TD technology have had most impact. Monitoring pressures during the various stages of system operation, SecureTD-Q (quantitative re-collection of splits during both primary and secondary desorption) and electronic mass flow control of split and column flows have all made an invaluable contribution to the issue of validation of desorption efficiency. Key examples of recent TD innovations and their application to the validation of sampling and thermal desorption efficiency will be described.
DESIGN CRITERIA FOR AN AUTOMATED CARTRIDGE SAMPLING SYSTEM FOR VOC CONCENTRATION AND FLUX MEASUREMENTS

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ABSTRACT

The design criteria of an automated constant flow sampler for sequential sampling of volatile organic compounds (VOC) on adsorbent cartridges are described. Features of the system include (a) the capability to collect up to 20 sequential samples to allow for long-term air quality assessments, (b) the ability to simultaneously collect two samples from different sources (e.g. for differential flux measurements), or to simultaneous collect two samples from one single source (e.g. for Quality Assurance backup samples), respectively, (c) to simultaneously collect on two different cartridge designs from one single source to increasing the range of analytical applications, (d) the use of inert wetted surfaces to ensure sample integrity, (e) the ability to set individual sampling times, intervals and flow rates by a microprocessor control unit with significant ease of operation, and (f) the capability to store data onto non-volatile memory during sampling for a complete audit trail of the sampling sequence. Various tests in the lab and during different field applications, including enclosure measurements, aerodynamic gradient techniques and relaxed eddy accumulation (REA) measurements, in a variety of environments (temperate and tropical climate) demonstrate the versatility of the automated VOC sampler. Low technical expenses, low power consumption, a robust weatherproof enclosure, and field portability allows sampling at remote sites and locations sensitive to disturbances or with restricted access.
AMMONIA MONITORING IN THE UK

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ABSTRACT

Ammonia (NH₃) emissions are estimated to have at least doubled over the last century across Europe, concomitant with intensification of agriculture, and increase in use of nitrogen fertilizers. The main sources of NH₃ in the atmosphere are from the decomposition and volatilisation of animal wastes. Other sources include direct volatilisation from synthetic fertilizers (particularly urea), and a wide range of non-agricultural sources such as sewage, catalytic converters, wild animals and industrial processes. It is recognised that deposition of atmospheric NH₃ contributes to acidification and eutrophication processes, which can cause damage to sensitive ecosystems.

The UNECE 1999 Gothenburg protocol and the EU National Emissions Ceiling Directive (NECD) of 2010 both set emission targets for individual countries. Abatement of NH₃ emissions is also included in EU Integrated Pollution Prevention and Control (IPPC) for the intensive pig and poultry sectors.

In the UK, the DEFRA-funded National Ammonia Monitoring Network (NAMN, www.edinburgh.ceh.ac.uk/cara/) was established in 1996 to quantify temporal and spatial changes in air concentrations and deposition in gaseous NH₃ and aerosol NH₄⁺ on a long term basis. The monitoring provides a baseline in NHₓ species, which is necessary for examining responses to changes in the agricultural sector and to assess compliance with targets set by international agreements. Data from the network are also used to test the performance of an atmospheric chemistry and transport model, FRAME that was developed at the same time with a special focus on NH₃.

There are currently 94 sites in the NAMN. At 57 of these sites, an active diffusion denuder methodology using the CEH DELTA (DEnuder for Long Term Atmospheric sampling) system is used to provide the main spatial and temporal patterns of NH₃ (and also NH₄⁺ aerosol) across the UK, whilst a high sensitivity passive diffusion sampler, the ALPHA (Adapted Low-cost Passive High-Absorption) sampler is implemented at a further 49 sites to assess regional and local scale variability in air NH₃ concentrations in source regions. To provide an ongoing validation of the ALPHA sampler, its performance is continuously assessed against the DELTA system at 12 sites within the network. The ALPHA sampler has also been tested in several international intercomparisons, e.g. EC ECOMONT project, and was included in the CEN TC264/WG11 pilot study into diffusive samplers for NH₃.
NEW APPLICATIONS FOR DIFFUSIVE SAMPLERS

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ABSTRACT

The decreasing sulphur dioxide levels in most parts of Europe and the increasing automobile traffic, causing elevated levels of nitrogen compounds, ozone and particulate matter have created a new multi-pollutant situation. Very few long-term measurements of atmospheric HNO₃ concentrations have earlier been performed despite the fact that HNO₃ is acidifying, eutrophying and causes corrosion and degradation on many materials. Long-term measurements in Sweden have shown a tenfold decrease of the SO₂ concentration during the last two decades, but no decreasing trend for nitric acid. The two gases now have equal concentrations on a molar basis

Atmospheric corrosion is a long-term effect, the relevant research therefore does not require monitoring on a daily basis. Moreover, power supply is often not available at sites where it is of interest to study the corrosion rate of objects belonging to our cultural heritage. Besides, such measurements must not disturb the impression of the objects.

Passive, long-term integrative techniques are ideal for sampling around objects of cultural heritage because the equipment is noiseless, don't need electricity and can be performed inconspicuously and with discretion. Other advantages are that technical personnel is not needed for exposing the samplers, the samplers don't need field calibration and the measurements are made in situ, i.e. reactive gases such as HNO₃ are not lost in inlet tubings. A diffusive sampler for HNO₃ has been compared to denuder technique in Gothenburg, Rome and Athens and has then been used to monitor concentrations at 11 rural and 23 urban widely spread sites over Europe.

Corrosion and degradation of objects belonging to our cultural heritage also takes place inside museums. Only about 5 – 10 % of the objects in a museum are displayed. The rest are stored in small boxes or storage rooms that often lack ventilation and the objects are inspected very seldom. The concentration of organic acids such as formic and acetic acid can be very high in closed rooms. The exhibited objects are also often stored in closed display cases. A diffusive sampler can be used to measure the emission of organic acids in a small closed compartment when the volume of the compartment is much smaller than the uptake rate of the sampler multiplied with the exposure time.

Eruptions are not the only problem with volcanoes. Mount Etna is the largest single source for SO₂ in Europe. The geographical concentration distribution of HF, HCl and SO₂ has been monitored in order to demonstrate the usefulness of diffusive sampling.

Even though we spend most of our time indoors, most directives concern the outdoor air, where also most air quality measurements are performed. Indoor air was in fact the first application for diffusive samplers. Today, diffusive samplers are available for more gases and vapours and better methods are available for evaluating the results. A new diffusive sampler for elemental mercury vapour has successfully been tested in the laboratory.

References
A CONTROLLED ATMOSPHERE TEST FACILITY FOR TRACEABLE MEASUREMENT OF UPTAKE RATE WITH VALIDATED UNCERTAINTIES

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ABSTRACT

The NPL Controlled Atmosphere Test Facility is designed to test and calibrate field samplers over wide ranges of ambient temperature, humidity and air speed. Multiple test gas mixtures of up to thirty adjustable component concentrations can be generated with traceability to national standards and with validated uncertainties. Uptake rates may be measured in laboratory-generated, test gas atmospheres and controlled environments that can be adjusted to resemble ambient conditions in the field. This allows realistic uptake rates to be determined, even in unusual atmospheres and ambient conditions and gives improved confidence in the quality and accuracy of air quality measurements. Pumped samplers can also be dosed by extracting test atmosphere from the facility.

The performance of the facility exceeds the requirements of EN 838, allowing the ambient conditions to be better controlled and defined to ensure uniform dosing of each of the exposed diffusive samplers. This reduces the contribution that variations in sampler dosing can make to the uncertainty in their measured uptake rate.

Accurate analysis and traceable calibration artefacts are essential in determining the amount of determinand collected by diffusive samplers, but without accurate uptake rates in applicable conditions the concentration at the sampling point cannot be derived with certainty. The NPL Controlled Atmosphere Test Facility is available to manufacturers and users of diffusive samplers to determine uptake rates, with a defined uncertainty, in selectable ambient conditions and atmospheres.
THE MANAGEMENT & COMMUNICATION OF AIR QUALITY DATA IN THE TYNE & WEAR REGION.

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ABSTRACT

Many local authorities monitor ambient air quality within their regions and generate a large archive of information. However, time constraints within local government have historically meant that much of this data is only used for the preparation of the reviews and assessments of air quality required by central government. Often these reviews and assessments are carried out by individual authorities (either in-house or by consultants) and fail to address regional issues that contribute to poor air quality.

It is also a requirement of both legislation and policy that the public are informed as to the quality of air in their regions and local authorities are increasingly using the world wide web for the dissemination of information on pollutants.

This paper will describe a project developed in the North East of England whereby both the database of information and the web site used to disseminate the pollution data, collected by five local authorities, is managed by Sunderland University.

Real time information on: nitrogen oxides, airborne particles (PM$_{10}$), sulphur dioxide, carbon monoxide and ozone is collected across the region by the five local authorities in the Tyne and Wear region. This information is polled by the University with daily checks being made on the quality of the data collected. At this stage each local authority is informed of any problems with a monitor in their region. Full reports on the data giving a regional perspective and the relationship of poor air quality to local meteorology are produced every three months. An annual report is also produced which is in a format that each local authority can submit to central government.

The web site is automatically updated daily and quality checks on the data are made by the University. Downloadable information on air pollution are both written and uploaded on to the web site by the University.
USE OF DIFFUSIVE SAMPLERS TO DETERMINE INDOOR AIR QUALITY AND VENTILATION

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ABSTRACT

Diffusive samplers are widely used for ambient air monitoring particularly for BTEX, NO₂, O₃, NH₃ and S0₂. Their usefulness for providing cost effective measurements of long term mean concentrations is equally applicable in indoor environments. Issues of method performance and appropriate sampling strategies are important for both indoor and ambient environments. For non-occupational indoor environments there has not to date been a strong legislative driver for development of methods, as in the ambient air, to verify compliance with air quality standards. However there has been sufficient scientific interest combined with the possibility of future requirements, and this has driven the preparation of standards in CEN and ISO for the diffusive measurement of some pollutants such as VOCs and formaldehyde and the measurement of ventilation. Examples include ISO 16017-2, ISO 16000-4, ISO DIS 16000-8 and EN14412.

An increasing number of countries including the UK have published air quality guidelines expressed as maximum concentrations for a defined exposure period. These are additional to recommended guidelines for indoor and ambient air for some pollutants published by the WHO. Air quality guidelines are being incorporated into Building Regulations (England and Wales) that determine ventilation, and performance targets for several pollutants are included in the revised ADF (Ventilation) that will be effective in 2006. This interest is due to recognition that indoor air quality is important for the health and well being of occupants, being the most important microenvironment for population exposure to many air pollutants, and this air quality can impact on productivity at the workplace and learning effectiveness in schools. The inhalation of pollutants indoors is also identified as a potentially important source to receptor link within contaminated land legislation.

Diffusive samplers have been applied by BRE as part of a wide range of studies of air quality in homes, offices and schools and to measure ventilation concurrent with these measurements. These studies show the usefulness and some current limitations of diffusive samplers for the study of indoor environments.

References


ABSTRACT

For a thermal desorption analysis to be successful, the adsorbent inside each tube and secondary trap must be properly packed. Any voids caused by settlement or contraction of the packing may induce channeling of the gas flow during pumped sampling and thermal desorption. Such effects may also disturb the geometry of the ‘air gap’ which is critical for predictable performance in diffusive sampling. Furthermore, the adsorbent may be damaged during improper packing, rough handling or by thermal shock giving rise to small fragments (fines) that fill the interstices between the packing particles and so serve to partially block the flow of gas during sampling and desorption. Clearly, tubes and traps that are correctly packed and maintained will give better sampling and analytical performance and should generate better data as a result.

In the case of glass tubes and traps, the condition of the packing may be assessed by visual inspection. This is not an option for diffusive monitoring because of the (opaque) metal tubes normally employed and so some other means must be found. One convenient technique to assess packing integrity would be to measure pressure drop across the tube or trap while a fixed flow rate of gas is applied. While such measurements have been previously performed manually, in this work we have adapted technology available on modern thermal desorption instrumentation to automate this procedure for both tubes and traps.

A fixed pressure of carrier gas is applied to the inlets of the tube or trap and mass flow controllers on the outlets are used to ensure that a fixed flow rate of gas passes through the packing. The pressure drops across both the tube and trap are recorded simultaneously and at the start of each analysis, but prior to desorption, using sensitive differential pressure transducers. To correlate the results for the tubes using the automated method against data from manual methods, the measured pressure drop is corrected to standard ambient temperature and pressure (SATP) using Darcy’s Law and the Poiseuille-Hagen equation. The application and performance of this new technology demonstrate that the automated measurement of packing impedance provides a useful measure of the condition of that packing for operational QC and diagnostic purposes.