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#### **1** Introduction

As the awareness of the correlation between airborne ultrafine particles and their potential threat to human health grows [1], surveillance of working areas with focus on particle emissions is becoming more and more important. The emission of soot particles from diesel combustion is receiving special attention, owing to the fact that large numbers of diesel engines for transportation and heavy-duty tasks are employed in various types of industry all over the world. While certain working areas are particularly endangered due to the lack of fresh or at least filtered air (e. g., construction sites in tunnels and mines), workers can also be exposed to high levels of particle emissions in closed buildings or other covered areas where ventilation is insufficient.

In many developed countries, occupational health is treated with great concern, and mandatory threshold limit values for the amount of foreign particle mass per unit volume of ambient air at working areas are being tightened. In the course of this development, the respirable ultrafine fraction of airborne particles is in the center of interest.

In the following, we present a simple and online method for continuous field measurements of the particulate elemental carbon concentration in air, stemming from diesel exhaust, at two selected working areas where occupational health is at risk.

#### 2 Experimental methods

2.1 Conventional detection of elemental carbon particles

The main constituent of particles from diesel engine emissions is elemental carbon (EC). A conventional and standardized method for determination of the EC content in air is coulometry [2], where a defined volume of air is sucked through a specially conditioned filter. The particulate fraction of the air volume is thereby deposited on the filter. Subsequently, the filter is chemically analyzed to obtain the respective EC content. Filter analysis methods like coulometry have several disadvantages: the results represent an integral value over a certain time period, the sampling times for

Dipl.-Phys. Karl Przybilla, Laboratorium für Festkörperphysik, ETH-Hönggerberg, Zürich (Schweiz). Dr.-Ing. Wolfgang Berkhahn, EcoChem Messtechnik, Überlingen. Prof. Dr. Heinz Burtscher, FH Aargau, Windisch (Schweiz). Dr. rer. nat. Dirk Dahmann, Institut für Gefahrstoff-Forschung – IGF der Bergbau-Berufsgenossenschaft, Bochum. Dr. Ulrich Matter, Matter Engineering, Wohlen (Schweiz). Dr.-Ing. Peter Rietschel, Berufsgenossenschaft Nahrungsmittel und Gaststätten, Mannheim. **Abstract** We present field measurements of diesel particulates at two different working areas with high loadings of diesel engine emissions. The concentration of airborne ultrafine elemental carbon particles (diameter below 1  $\mu$ m) is measured with a photoelectric aerosol sensor. Comparison of the sensor signal with the results of coulometric filter sampling, done in parallel at some locations, show that the photoelectric aerosol sensor is well-suited as a simple and online measuring device, especially for monitoring the particulate emissions of diesel engines. Laboratory experiments confirm the linearity and long-term stability of the sensor signal.

### Einsatz des photoelektrischen Aerosolsensors zur Überwachung der Diesel-Partikelkonzentration an Arbeitsplätzen

**Zusammenfassung** Feldmessungen zur Bestimmung der Konzentration von Ultrafeinpartikeln aus elementarem Kohlenstoff (Durchmesser kleiner 1 µm) wurden an zwei verschiedenen Arbeitsplätzen mit hoher Belastung durch Dieselmotoremissionen durchgeführt. Der Vergleich des Messsignals eines photoelektrischen Aerosolsensors mit Resultaten aus Coulometrie-Filteranalysen zeigt, dass sich der photoelektrische Aerosolsensor als Online-Messgerät besonders zur Arbeitsplatzüberwachung hinsichtlich partikelförmiger Dieselmotoremissionen eignet. Laborexperimente mit Kohlenstoffpartikeln bestätigen die Linearität und die Langzeit-Stabilität des Messsignals.

sufficient filter loading are usually quite long, and the analysis has to be done in specially equipped laboratories which makes the whole process time consuming and expensive.

Coulometric analysis was performed according to [2]. The filter samples for coulometry were taken according to [3] on a MPG II (pre-separator: horizontal elutriator) or on a PM4 F (pre-separator: cyclone) fine dust measuring and sampling instrument. Both devices are equally well suitable and interchangeable.

#### 2.2 Online detection of elemental carbon particles

A simple and online method for the detection of small EC particles in air is by means of photoelectric charging. A photoelectric aerosol sensor (PAS) takes continuous samples of ambient air and irradiates the sampling volume with UV light. Compared to other airborne particles, the surface of EC particles exhibits a workfunction that is low enough to permit emission of photoelectrons upon irradiation with UV light. If the particles are sufficiently small (diameter <  $1 \mu m$ ), the probability for reattachment of photoelectrons to the particle is small as well. In this case, the particles remain positively charged and can be measured electrically [4]. It has been shown in many experiments with particle emissions from diesel engines that there is a good correlation between the measured signal from the PAS and the EC content as detected by means of coulometric filter analysis [5; 6]. Owing to this fact, the PAS is suitable for online monitoring of the ultrafine EC particles emitted from diesel combustion processes, permitting a time resolution of up to 1 second. A PAS enables to perform constant, automatic, and time-resolved surveillance of working areas with respect to the concentration of ultra-



Figure 1 Ground plan (schematic) of the indoor loading area and the block storages of the brewery. The points denominated MP indicate the locations where air quality measurements were performed.

fine EC particles in air. The PAS model used was a PAS 2000 by EcoChem.

#### **3 Field measurement 1: Bitburger Brewery, Germany**

#### 3.1 Motivation and purpose of the measurements

The Bitburger brewery in Bitburg, Germany, operates an indoor loading area for the purpose of unloading empty goods from and loading full goods onto diesel trucks. These tasks are performed by approximately 70 employees in three shifts by means of diesel fork-lift trucks (model Linde H 80 D, 8 tons lifting capacity, equipped with soot particle filters). The goods are taken from and brought to block storages that are situated sideways of the loading area. Work is organized in two shifts per day-time and an additional shift at night-time with a reduced number of employees.

The performed measurements are motivated by the fact that the diesel soot emissions inside the indoor loading area are increasing steadily and the existing ventilation system no longer meets the demands with respect to workplace air quality. Though the fork-lift trucks are equipped with soot particle filters, it is assumed that the diesel soot emissions inside the loading area mostly stem from them because they operate much more than the trucks entering and leaving the loading area only. The brewery is planning to install a new and efficient ventilation system, based on insights resulting from measurements performed on the existing ventilation system.

#### 3.2 Measurement program

The indoor loading area is equipped with 48 ventilators (model LTG VRG710). Each ventilator can be operated either for removal of 12 800 m<sup>3</sup>/h exhaust air only, or as a combined 5 300 m<sup>3</sup>/h fresh air supply and 6700 m<sup>3</sup>/h exhaust air ventilator with heat recovery. The block storages are equipped with fresh air supplies only. The dimensions of the loading area are 325 m (length), 30 m (breadth), and 10 to 12 m (height).

#### 3.2.1 Test run 1: ventilation at half power

For this measurement, only 12 exhaust air ventilators of the first half of the loading area are operated (50 % ventilation power), leading away 150 000 m<sup>3</sup>/h of exhaust air. The necessary additional air is supplied by 12 ceiling ventilators in the block storage which provide a volume flow of 120 000 m<sup>3</sup>/h. The additional air enters the indoor loading area mainly near floor level. This set-up leads to an air exchange rate of 2.6/h. Both online PAS signal and EC content by means of coulometric filter analysis are measured in the centre of the first half of the loading area.

3.2.2 Test run 2: ventilation at full power

### For this measurement, all 24 exhaust air ventilators of the first half

of the loading area are operated (100 % ventilation power), leading away a total of 300 000 m<sup>3</sup>/h of exhaust air. The lack of fresh air compared to the first test run is, among other things, additionally supplied by an open door in the block storage (15 000 m<sup>3</sup>/h) and a partially opened gate in the middle of the first half of the loading area (43 000 m<sup>3</sup>/h). This arrangement leads to an air exchange rate of 5.3/h. Again, both online PAS signal and EC content by means of coulometric filter sampling are measured in the center of the first half of the loading area.

#### 3.3 Evaluation of the ventilation system

From the first two test runs a conversion factor between PAS signal and EC content (coulometry) is obtained. The PAS signal can now be translated directly into the respective EC concentration. With this information it is possible to perform measurements at different locations inside the indoor loading area with the PAS and convert the mean PAS signal into the corresponding EC concentration. Thus, a spatially resolved analysis of the ventilation system performance in the whole loading area can be undertaken. Different measurement locations inside the loading area are examined, whereby measurements are taken both at floor and at ceiling level (**Figure 1**).

#### 3.4 Results and discussion

Figure 2 shows the online signal of the PAS together with the respective EC value as determined by coulometry inside the first half of the loading area for ventilation at half and full power (test runs 1 and 2). The sampling time lies between one and a half and two hours. Comparison of the mean PAS value with the EC content, determined by means of coulometry, leads to a conversion factor that can be used to translate the measured PAS current into the corresponding amount of EC mass per m<sup>3</sup>. For better comparison, the coulometric EC value is multiplied by a factor of 10 000. It has been shown in various measurements on diesel engine emissions that the mean PAS signal is proportional to the integral value of the EC concentration as determined by coulometry [5; 6]. The results from Figure 2 are in good agreement with these findings. It follows from the measured data that the EC concentration in  $mg/m^3$ , as determined by coulometric filter analysis, can be derived from the online PAS signal by dividing the mean PAS

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signal by 10 000. This conversion is used to evaluate the EC concentration by means of online measurements with the PAS at different locations inside the loading area.

**Figure 3** shows the results of various PAS measurements at different locations inside the loading area. At each measuring point (MP) presented, the online PAS signal was recorded for 10 minutes. With the conversion factor obtained from the first two test runs, the mean PAS signal can be directly translated into the EC concentration in mg/m<sup>3</sup> at the respective measuring point. This has been done for the data presented in Figure 3.

MP 6 is located in the first half of the loading area, close to one of the ventilators. The EC concentration at ceiling level is roughly double as high as at floor level. This is a consequence of three factors: the fresh air enters near the floor level, the fork-lift trucks emit their exhaust gas upwardly, and the hot exhaust gases are driven to the ceiling by convection.

MP 7 is also located in the first half of the loading area. In contrast to MP 6 it lies in a segment of the ceiling without a ventilator. The EC concentration at floor level is about the same as for MP 6. The EC concentration at ceiling level, however, is three times higher than at floor level. This can be explained by the fact that there is no ceiling ventilator close to this measuring point. The exhaust gases accumulate in this segment of the ceiling.

MP 9 is situated in the second half of the loading area. As opposed to the first half where all ventilators are driven in exhaust air mode only, the ventilators in the second half are operated in the mixed fresh air/exhaust air mode, where both fresh air and exhaust air are collected and emitted through the ceiling. Under the mixing mode condition, the EC concentration at floor level is higher by 0.025 mg/m<sup>3</sup>, as compared to MP 6. At ceiling level, however, the EC concentration is lower by 0.150 mg/m<sup>3</sup>, as compared to MP 6.

MP 10 is located in an adjacent hall to the first part of the loading area. There was no traffic at the time of measurement. In this hall, fresh air is supplied from the ceiling. This results

in a comparatively low EC concentration at ceiling level. At floor level, however, the EC concentration is elevated. This may be related to an effect that emissions settle to the floor while fresh air is supplied from above.

These data have greatly assisted the evaluation of the existing ventilation system within the loading area of the Bitburger brewery and moreover the design of a new system. They show clearly that it is necessary to perform air quality measurements at various locations across the indoor loading area in order to ensure that a new ventilation system can be designed properly in the sense that the required limiting values for the EC concentration are not exceeded anywhere inside the loading area. In particular, the measurements have shown that in this case a ventilation with air supply from



Figure 2 Measurement of the online PAS signal and the corresponding EC concentrations (horizontal lines), as measured by coulometric filter analysis, for ventilation at half and full power. For better comparison, the EC values are multiplied by 10<sup>4</sup>.



Figure 3 Online and mean PAS signal at different locations inside the indoor loading area. The PAS signal has been converted into the corresponding EC concentration in units of mg/m<sup>3</sup>. The solid line indicates the mandatory threshold limit value of 0.1 mg/m<sup>3</sup> at working areas according to [7].

floor level and air extraction at the ceiling seems more suitable than a mixing ventilation. Due to the online character of the PAS data, a fast assessment of the EC concentration throughout the whole loading area is feasible. If necessary, continuous surveillance of the air quality is possible.

The EC concentration inside the loading area obviously depends on the amount of traffic, i. e., on the amount of trucks being loaded at the same time. It is worth mentioning that a permanently installed PAS could provide data for the steering unit of the new ventilation system, allowing to adjust the ventilation power automatically according to the momentary air contamination.



Figure 4 Measurement of the online PAS signal and the corresponding EC concentrations (rhombs), as determined by coulometric filter analysis, inside the tunnel construction site. The horizontal line indicates the mandatory threshold limit value of 0.1 mg/m<sup>3</sup> at working areas according to [8].



Figure 5 Correlation between mean PAS signal and the EC concentration as measured by means of coulometric filter analysis.

## 4 Field measurement 2: railroad tunnel construction site, Switzerland

#### 4.1 Motivation and purpose of the measurements

The Swiss Federal Railways (SBB) are constructing a new tunnel in Thalwil near Zurich in order to cope with the growing volume of traffic to and from the city of Zurich. For the purpose of evaluating the workplace conditions in a tunnel construction site, air quality measurements with emphasis on the EC concentration were performed inside the tunnel construction site.

#### 4.2 Measurement program

An online PAS was placed next to a coulometry sampling point where filter samples of the air inside the tunnel were taken in hourly intervals. The tunnel has a length of 4 600 m and is ventilated by means of three air tubes with a diameter of 180 cm, transporting between 18 and 60 m<sup>3</sup> of fresh air per second into the tunnel. The exhaust air flows back outside through the main tunnel with a velocity of 1.06 m/s. Work is organized in two shifts between 06:00 and 22:00 o'clock. While the highest particle emissions obviously occur at the main construction points, namely in two sidewing tunnels that lead away from the main tunnel, the PAS and coulometry samples where taken in the main tunnel closer to the tunnel exit in order to examine the contamination of the backflowing air.

Tunnel construction consists of a multitude of different worksteps. Among diesel particle emissions from heavy-duty drilling machines and transporting trucks, the tunnel air is also filled with a lot of other foreign matter like dust resulting from blasting events and shotcrete treatments. The humidity, in general, is elevated inside the tunnel.

#### 4.3 Results and discussion

Figure 4 shows the measured online PAS signal for selected measurement intervals inside the tunnel. The big fluctuations are due to the different worksteps performed on the day of measurement. The integral EC concentration as measured by coulometry is also displayed, together with the legally allowed threshold limit value of EC for diesel engine emissions  $(0.1 \text{ mg/m}^3)$ . It is visible that the mandatory limit value is clearly exceeded at certain points in time. In the case of the last measurement cycle, for example, even the integral coulometric EC value exceeds the threshold limit value. From Figure 5 it can be seen that, also for the aerosols inside a tunnel construction site, there is a good correlation between the PAS signal and the EC concentration as measured by coulometry. It is, therefore, again possible to calculate a conversion factor for the PAS, allowing to translate the PAS signal directly into the respective EC mass concentration. In the presented case, a division

of the PAS signal by 7 leads directly to the respective EC mass concentration in units of  $\mu g/m^3$ .

#### 5 Laboratory measurement: graphite particles

#### 5.1 Motivation and purpose of the measurements

The field measurements have shown that, in both cases, there is a good correlation between the signal of the PAS and the EC mass content as measured by coulometry. While the conversion factor depends on the type and origin of the aerosol, the correlation between the two measures is conserved over a wide range of particle concentrations. This property is also confirmed in laboratory experiments, where the particle composition is better known and where the particle concentration can be varied according to requirements.

#### 5.2 Measurement program

Carbon particles in the nm size range are produced by a Palas GFG-1000 spark discharge particle generator with graphite electrodes. The particles are formed and carried in a nitrogen gas flow in order to minimize contamination of the particle surface. A certain fraction of the carbon particles is extracted by means of a rotating disk dilution system [9]. In so doing, different number concentrations of carbon particles can be fed into the measuring equipment while the particle size stays unchanged.

The carbon particles are fed in parallel into a PAS and a scanning mobility particle sizer (SMPS) [10], a device which classifies and counts the particles according to their electrical mobility diameter.

This experimental arrangement serves two purposes. First, the linearity of the PAS signal with respect to the particle concentration is verified. Second, the long-term stability of the PAS is examined.

#### 5.3 Results and discussion

In order to examine the linearity of the PAS with respect to the particle number concentration of a defined aerosol, various number concentrations of carbon particles were fed into the sensor by means of a dilution unit. Shape and electrical mobility diameter of the carbon particle spectrum were checked regularly in parallel with the SMPS system. In **Figure 6**, the PAS signal and the total particle number concentration are plotted as a function of the dilution ratio. The shape of a typical size spectrum is also displayed (inset).

The linearity of the PAS signal as a function of particle number concentration is good. Measurements with carbon particles were conducted with various particle sizes (shift of the particle spectrum). This had no significant influence on the linearity of the signals.

In order to examine the reproducibility and long-term stability of the PAS, a defined size spectrum of carbon particles was generated on numerous days. The constancy of number concentration, average mobility diameter, and shape of the size spectrum were verified regularly with the SMPS system. In **Figure 7**, both mean values of the PAS signal and the overall mean value are plotted over a time period of six months. The shape of the particle distribution is the same as in Figure 6. The measurement reproducibility of most aerosol measuring techniques is usually in the range of some ten percent. In comparison, the presented result is quite satisfying and shows that the PAS offers reproducible results, especially over a long period of time.







Figure 7 Long-term test of the PAS 2000 on a constant aerosol source (carbon particles).

#### 6 Conclusions

It has been shown in two different field measurements that there is a good correlation between the measuring signal of a PAS and the EC concentration as measured by coulometry. It is possible to obtain a conversion factor that allows to translate the online signal of the PAS directly into the actual EC concentration. Once this information has been acquired, the PAS emerges as a simple, fast, and cheap device for determination of the EC concentration in air. The PAS yields an online signal with a time resolution of up to 1 second, allowing to observe rapid changes of the ambient EC concentration. It measures continuously and is therefore well-suited for online surveillance of respirable ultrafine carbon particles at working areas.

The general properties observed in field measurements have also been analyzed in laboratory experiments under well-controlled conditions. The linearity of the PAS signal as a function of the particle number concentration could be shown clearly and for different particle sizes below 1  $\mu$ m. Additionally, the reproducibility and long-term stability of the PAS could be demonstrated.

It has to be pointed out that, while there is generally a good correlation between the PAS signal and the EC concentration from particles in air, the respective conversion factor depends on the type and origin of the aerosol that is being analyzed. This is related to a certain cross-sensitivity of the photoelectric charging method with regard to the respective particle surface composition. However, as long as the chemical composition of the measured particles does not change significantly, the PAS is qualified for reliable monitoring purposes of the ambient air. If the aerosol composition is unknown or changes fundamentally, the consistency of the acquired PAS data should be cross-checked with results from other sources, e.g., coulometry.

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Herausgebermitteilungen

### Aus der Arbeit der KRdL

VDI 2310 Blatt 6: Maximale Immissions-Werte zum Schutz der Vegetation – Maximale Immissions-Konzentrationen für Ozon. Juni 2002. Ersetzt den Entwurf vom Dezember 2000. Erscheint in deutsch/englischer Fassung. 152,30.

Die Richtlinie VDI 2310 Blatt 6 behandelt maximale Immissions-Konzentrationen für Ozon zum Schutz der Vegetation. Infolge umweltpolitischer Maßnahmen ist in den letzten Jahren eine Veränderung der Belastungssituation von Mensch (siehe auch VDI 2310 Blatt 15, Dezember 2001) und Umwelt eingetreten. Spitzenbelastungen durch bodennahes Ozon konnten zunehmend reduziert werden, so dass ein Trend hin zu mittleren Belastungen erkennbar ist. Diese sind jedoch hinsichtlich der Gefährdung der Vegetation wirkungsbestimmend, da extrem hohe Ozon-Konzentrationen meist mit Bedingungen korrelieren, die ein Auftreten von Schäden infolge z. B. niedriger Aktivität der Pflanzen (Gasaustausch) ausschließen.

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Derzeitige Europäische Grenzwertkonzepte zur Einschätzung der Gefährdung der Vegetation führen aufgrund der eingeschränkten Datenlage und verschiedener Unsicherheiten zu z. T. überhöhten Schädigungsprognosen. In der VDI-Richtlinie wurde daher eine Sichtung des aktuellen Datenmaterials aus europäischen Erhebungen vorgenommen und in Dosis-Wirkungs-Beziehungen für Wild- und landwirtschaftliche Nutzpflanzen sowie Bäume übertragen. Die unter Berücksichtigung eines flussorientierten Ansatzes für verschiedene Zeiträume ermittelten Maximalen Immissions-Konzentrationen für Ozon beziehen sich auf die tatsächlich toxikologisch wirksame Ozon-Konzentration in Pflanzenhöhe, so dass nur eine Umrechnung der in der Referenzhöhe gemessenen Ozon-Konzentration zu realistischen Abschätzungen führt. Die Notwendigkeit des Einsatzes flussorientierter Ansätze zur Abschätzung der tatsächlichen Gefährdung der Vegetation ist akzeptiert und findet erstmalig in dieser Richtlinie Anwendung.

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