

THE CORRELATION BETWEEN IN-SITU, REAL-TIME AEROSOL PHOTOEMISSION INTENSITY AND PARTICULATE POLYCYCLIC AROMATIC HYDROCARBON CONCENTRATION IN COMBUSTION AEROSOLS

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ABSTRACT. Aerosol photoemission (APE) has been found to be an excellent surrogate measure of particulate PAH concentration in combustion aerosols under a variety of laboratory and field conditions. Samples of oil stove, automobile exhaust, and urban air particulate matter were concurrently analyzed for aerosol photoemission intensity and particulate PAH concentration. In-situ, real-time analysis of the particulate matter was performed by UV-induced electron photoemission. Two photon energies, 4.9 and 6.7 eV, were chosen for irradiation. The latter energy was more sensitive and less selective. Particulate PAH concentrations were determined by collecting the particulate matter on a filter, solvent extracting the filter, and analyses of the extracts by gas chromatography / mass spectroscopy. Linear correlations ($0.84 \leq r^2 \leq 1.00$) were observed between aerosol photoemission intensities and the sum of particulate PAH concentrations for laboratory and field studies under a wide variety of conditions. The studies performed to date cover a dynamic range of three orders of magnitude (1 to 1000 ng / m³). The time resolution is in the order of a few seconds.

1. Introduction

The atmosphere plays a major role in the transport and cycling of natural and anthropogenic organic chemicals. Atmospheric inputs into ecosystems can be broken down into its two primary inputs; inputs from the gas and from the particulate phase. The extent to which compounds are removed from the atmosphere, and become available to other compartments, depends in part on how a chemical is distributed between the gaseous and particulate phase. Total suspended particulate matter (TSP), especially that associated with incomplete fossil fuel combustion, is an important vehicle for transport of the relatively non-volatile, high molecular weight organic compounds in the environment (Bidleman, 1988) and in the human respiratory tract (Heinrich, 1989). The carcinogenic properties of particulate matter from incomplete fossil fuel combustion sources have been identified as early as 1775 by Sir Percival Potts (Bayland, 1983), who identified the relationship between the high incidences of testicular cancer with the chimney sweep profession. This led to numerous studies examining the carcinogenic properties of incomplete fossil fuel extracts (e.g. Passey, 1922). Goulden and Tripler (1949) were the first group to identify a specific polycyclic aromatic hydrocarbon (PAH), benzo(a)pyrene, in soot and determine that it was one of the compounds responsible for the high carcinogenic activity. These results launched numerous studies over the next 40 years for the determination of the concentrations and subsequent health effects of PAHs in particulate matter as a result of incomplete fossil fuel combustion.

The determination of the concentration of PAHs in TSP is typically a four step process: 1) sampling and collection of TSP; 2) extraction of the PAHs from the TSP; 3) concentration of the PAHs in the extracts; and 4) analytical determination of the PAHs in the concentrated extracts. The sampling and collection of TSP is an extremely difficult process that is fraught with many potential sources of error associated with artifact formation. Ideally, the collected TSP would represent a "snapshot" of what is actually present in the atmosphere. However, in reality, this is not always the case. Samples of TSP are typically collected by drawing ambient air through a fibrous or membrane filter (Bidleman, 1985; McDow, 1986; Hart, 1989). The three major artifacts associated with this technique are volatilization of collected TSP, chemical reactions on the collected TSP, and gas phase adsorption to the filter (Lee and Schuetzle, 1985). If sampling artifacts are present, the first two will tend to underestimate the concentration of a compound in the particulate phase and the last one will have the opposite effect. Diffusion denuders have also been developed to obtain potentially artifact-free samples of TSP (e.g. Appel et al., 1983). However, both types of samplers have experienced limited success and are only concerned with the first consideration in the determination of a compound concentration in TSP, i.e. TSP collection.

The second step, the extraction of PAHs from TSP, is also a very complicated procedure that is dependent on a number of physical and chemical characteristics of the TSP and has been reviewed by a number of authors (Griest and Caton, 1985; Grimmer, 1985; Lee and Schuetzle, 1985; McDow et al., 1990). While the extraction and concentration steps outlined above are tedious, time intensive, and expensive to perform on a routine basis, research level solutions have been addressed in numerous studies (e.g. Giger and Schaffner, 1978; Van Vaeck and Van Cauwenberghe, 1978; Griest and Caton, 1985; Hart et al., 1989; McDow et al., 1990). Although the analytical determination of the PAHs in the extracts is relatively straightforward by gas chromatography / mass spectrometry or high performance liquid chromatography, the capital expenses associated with purchasing and upkeep of these instruments are relatively high. Another potential difficulty using this technique is that in order to collect enough TSP for quantitation using standard high-volume samplers, the minimum practical time resolution is typically 4 to 24 hours. Any temporal perturbations with a timeframe less than the sampling time will be averaged over the whole sampling time. Thus, an in-situ, real-time technique that minimizes or eliminates the above mentioned potential difficulties of determining PAH concentrations in TSP is greatly needed.

An alternative method to the above approach for determining the concentration of PAHs in TSP has been developed using photoelectric charging of particles by UV-light (Burtscher et al., 1982; Niessner, 1986; McDow et al., 1990). This method, referred to as aerosol photoemission (APE; Burtscher et al., 1988) has the advantages of: 1) artifact-free, in-situ measurement of particulate matter; 2) an extremely sensitive signal when photoemitting surfaces (e.g. particulate PAHs) are present on particulate matter; and 3) real-time temporal resolution of a few seconds. For the APE technique, in-situ refers to the measurement of APE on TSP as it occurs in ambient air. This is in contrast to the methods that collect TSP on a filter and subject the TSP to the various sampling artifacts mentioned above. A review on photoelectric charging is given by Burtscher and Siegmann (1992) in this volume.

Burtscher et al. (1982, 1988) have measured high photoemission intensities from PAH laden particulate matter as a result of incomplete fossil fuel combustion in automobiles and other combustion sources. McDow et al. (1990) have shown that a significant positive linear correlation was found between the sum of particulate PAH concentrations and photoelectric charge densities with oil burning stoves in the laboratory and with TSP in parking garages. In this study particulate PAH concentrations and photoelectric charge densities of urban air in Zürich, Switzerland are measured. Based on these immission data and the data by McDow et al. (1990) this manuscript will address the usefulness and potential applications of using APE as a

surrogate method to measure the sum of the particulate PAH concentrations in the laboratory and field.

2. Experimental

The APE apparatus that was used in both studies is shown in Figure 1 and has been described in detail by Burtscher and Siegmann (1992) and Burtscher et al. (1988).

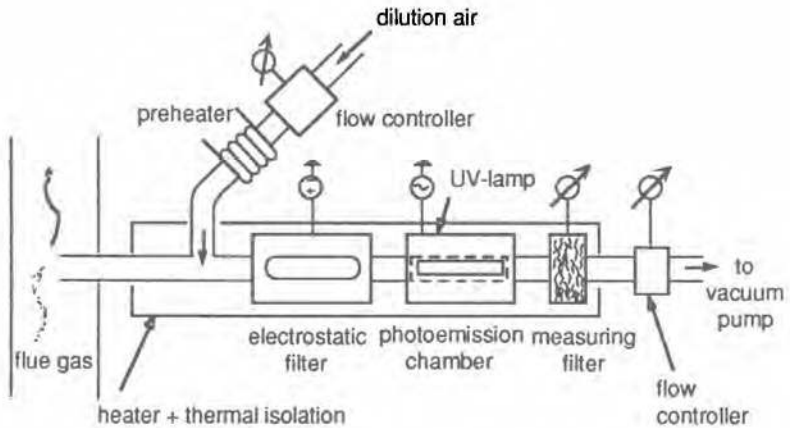


Figure 1. Aerosol photoemission (APE) apparatus. Dilution inlet only utilized during the collection of oil stove particulate matter.

McDow et al. (1990) used a light source of 4.9 eV photon energy, in this study a 6.7 eV lamp is used. As the sensitivity of the to different PAH's depends on the photon energy, a direct comparison of results is only allowed for identical PAH-profiles, which is not exactly the case. Nevertheless the two sensors were calibrated to give the same reading for automobile exhaust by parallel measurement in a car parking garage. This calibration is used for comparison of the data by McDow et al. (1990) with the data from this study. Though this comparison is problematic, it is justified or the PAH-profiles are similar.

McDow et al. (1990) used a 142 mm glass fiber filter at a flow rate of 48 m³/h for particle collection. In this study 20x25 cm quartz fiber filter were applied, the flow rate is the same. The sampling head that was used utilized two quartz fiber filters in series to collect TSP (Hart, 1989; Hart and Pankow, 1991). Since the collection efficiencies of TSP on quartz or glass fiber filters is extremely efficient (> 99.99%), the backup filter gives a measure of the amount of gas phase adsorption to a filter and allows the gas phase to be subtracted from the total PAH concentration measured on the primary filter. Volatilization from TSP collected on the filter surface was minimized in both studies by operating at high volumetric flow rates, thus minimizing the sampling time. Thus, fluctuations in ambient temperature, sampling different air parcels, and gas / particle partitioning were minimized.

Once the samples were collected, the filters were extracted, the extracts concentrated, and analyzed by gas chromatography / mass spectrometry (Giger and Schaffner, 1978; Lee and Schuetzle, 1985; McDow et al., 1990;). A total of 16 PAHs were analyzed and are referred to as particulate PAHs: phenanthrene, fluoranthene, pyrene, benzo(b)naphtho(2,1-d)thiophene, benzo(ghi)fluoranthene, benzo(c)phenanthrene, benz(a)anthracene, chrysene, benzo(b+j+k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, benzo(ghi)perylene, and coronene. Recovery and blank studies are included in the above references. The APE signal was averaged over the time period in which the samples were taken for GC / MS analyses.

3. Results and Discussion

In the laboratory, McDow et al. (1990) sampled exhaust gas that resulted from burning No. 2 fuel oil in an Oertli OE-9 burner and Strebel G14/4 boiler at different settings of the air inlet damper. They monitored the aerosol production at the beginning and end of each sampling period with a Bacharach test which visually compares the loading or "blackness" on a filter to a standard scale (DIN 51402, 1986). A Bacharach number of ≤ 1 indicates normal or typical burning conditions with optimal fuel / oxygen ratios that result in minimal aerosol production.

Figure 2 shows the sum of the particulate PAH concentrations (PPAH; calculated by summing the concentrations of all particulate phase PAHs with $178 \leq MW \leq 300$; McDow et al., 1990) as a function of Bacharach number.

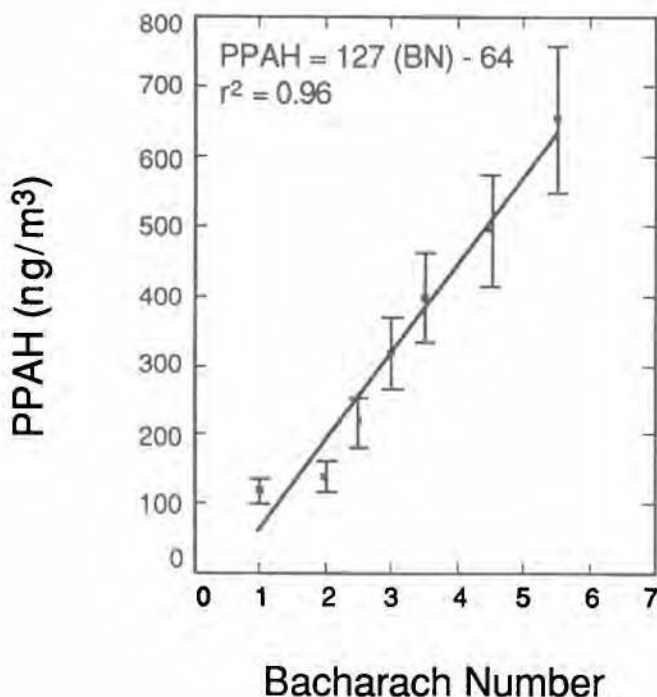


Figure 2. Bacharach number vs. PPAH concentration from particulate matter collected from the exhaust of an oil stove burning No. 2 fuel oil. Adapted from McDow et al., (1990). Error bars shown are ± 1 standard deviation.

By reducing the air supply to the oil burner, the amount of incomplete combustion increases, leading to increasing Bacharach numbers. Note that Figure 2 shows an order of magnitude increase in PPAH concentration as the Bacharach number increases from 1 to 6. A good positive correlation ($r^2 = 0.96$) was obtained by a least squares linear regression of the data in Figure 2. Error bars associated with each data point are ± 1 standard deviation based on standard propagation of error techniques. Using the extraction method outlined by Grimmer et al. (1982) with toluene as a solvent, the

PPAH concentration increased with increasing Bacharach number (McDow et al., 1990). These results are in contrast to a previous study by Leary et al. (1987), where the total PPAH concentrations were relatively constant with increasing Bacharach number. However, this apparent contradiction is most likely tied to different operating characteristics of the oil stoves and the extraction efficiencies of the solvents that were utilized in the studies (Griest and Caton, 1985; Grimmer, 1985; Lee and Schuetzle, 1985; McDow et al., 1990). Thus, for the conditions encountered in the McDow et al. (1990) study, the PPAH concentrations were strongly related to the degree of incomplete combustion as measured by the Bacharach number.

The correlation between PPAH from the oil stove described above and photoelectric charge density can be seen in Figure 3.

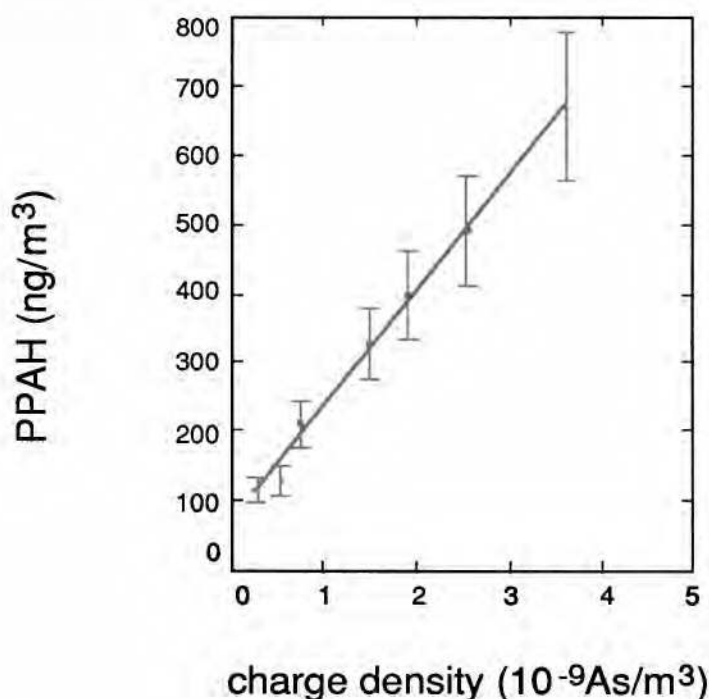


Figure 3. Photoelectric charge density vs. PPAH concentration from particulate matter collected from the exhaust of an oil stove burning No. 2 fuel oil. Adapted from McDow et al., (1990). Error bars shown are ± 1 standard deviation.

The regression parameters for all of the data presented in this manuscript are given in Table 1. An excellent positive correlation ($r^2 = 0.99$) can be seen for almost an order of magnitude increase in PPAH concentration. While the sampling protocol can not be directly compared in the laboratory oil stove study by McDow et al. (1990) and the field sampling of TSP by McDow et al. (1990) and this study, the photoelectric charge densities from the oil stove particulate matter were approximately two orders of magnitude higher than those of the field studies. The results presented in Figure 3 support the hypothesis that APE can be used as a surrogate measure to estimate PPAH concentration adsorbed on the surface of particulate matter.

TABLE 1. Regression parameters for particulate PAH concentration (PPAH; ng/m³) vs. aerosol photoemission (APE; As/m³) for laboratory and field measurements in Zürich, Switzerland. The regression equation^a is: PPAH = m(APE) + b.

Type	m (x 10 ¹¹)	b	r ²
Laboratory Oil Stove ^b	1.67		
Hönggerberg Parking Garage ^b	7.75	+5.90	0.84
Dübendorf (D)	7.73	-0.45	0.83
Zürich 1 (Z1)	5.66	-0.01	0.94
Zürich 2 (Z2)	7.04	-1.49	0.90
Z 1 & Z 2	6.09	-0.51	0.92
Z 1, Z 2 & D	5.67	+0.29	0.87
all data ^c	0.87	+10.41	0.93

a) m = slope, b = y-intercept; b) McDow et al., 1990;
c) log PPAH = log {m(APE)} + b; data as Figure 8.

The APE measuring technique was then taken into the field to examine the correlation between PPAH concentration and photoelectric charge density on TSP. The results from the McDow et al. (1990) study using TSP collected in a parking garage at ETH Hönggerberg and corresponding photoelectric charge densities are presented in Figure 4. While the correlation ($r^2 = 0.84$) was not as high as was seen for the oil stove data presented above, it should be noted that the samples were collected over a three day period with different spatial proximity to the sources. The TSP that was sampled may have aged to differing degrees depending on the meteorological conditions,

the residence time of the TSP in the garage, and the spatial proximity of automobiles and other sources. Another factor could be the influence of diesel engines, relative to gasoline engines. Stenberg (1985) has shown that PPAH extracts from diesel exhaust particulate matter have ~ six times as much PPAH as exhaust from gasoline engines. McDow et al. (1990) also noted that the characteristic PPAH profiles found in TSP (Grimmer et al., 1982; Grimmer, 1985), were quite different from the PPAH profiles found in oil stove extracts (Leary et al., 1987). These results are encouraging to the extent that such a good correlation was found between PPAH concentrations and photoelectric charge densities under the above conditions.

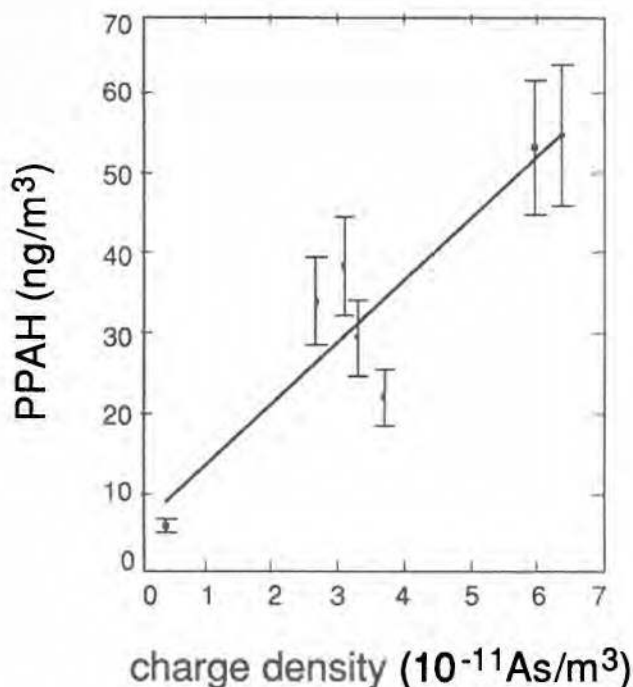


Figure 4. Photoelectric charge density vs. PPAH concentration from TSP collected from the parking garage at ETH Hönggerberg, 1988. Adapted from McDow et al., (1990). Error bars shown are ± 1 standard deviation.

Since the APE technique was successful in examining TSP collected at the ETH Hönggerberg parking garage, the applicability of using APE as a surrogate measure PPAH concentration was examined at two sites in and near Zürich, Switzerland (see Fig.5). As can be seen from Figure 5, Dübendorf lies

East of Zürich, and under typical summer meteorological conditions, is downwind of the industrial center of Zürich. Dübendorf is also adjacent to major freeways and roads, a commercial and military airport, and industrial complexes. Thus, TSP in an air parcel moving in an Easterly direction would experience a relatively large input of "fresh" TSP from Zürich and, to a lesser extent, from Dübendorf.

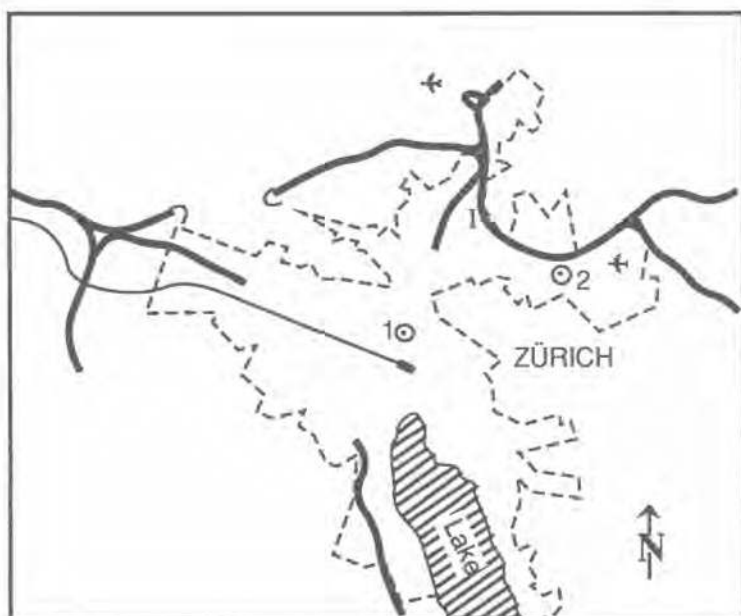


Figure 5. Map of sampling area in Zürich, Switzerland; Collection site 1: Zürich city center, 2: Dübendorf, I: waste incinerator. The bold lines are the main highways. For easier orientation the rail track from Zürich, main station (thin line) is also shown, though not important for emissions.

A total of 12 TSP samples were collected for PPAH determination while continuously measuring photoelectric charge density in Zürich, and Dübendorf. An example of photoelectric charge densities as a function of time is shown in Figure 6. An interesting aspect of this graph is the increase in photoelectric charge density at midnight as a function of the days of the week. The photoelectric charge density is approximately an order of magnitude higher on Friday than on Monday and only a factor of three higher than on Thursday. More measurements would be required to decide whether

this is really day specific and e.g. due to increased nighttime activity towards the weekend or caused by the meteorological conditions. Total suspended particulate matter (TSP), elemental carbon (EC), wind speed and direction, and a surface area were also measured in this study, and their correlations with photoelectric charge density are treated elsewhere (Burtscher et al., 1992). The measurements can be into three periods, each lasting 3 days: One at Dübendorf (May 21-23,1991), two in Zürich (May 27-29, June 4-6, 1991). A total of four samples were taken from each period. The sampling times were chosen to cover the morning, afternoon, and evening time slots along with a night sample, where photolytically catalyzed reactions on the APMs surface are low and inputs from incomplete fossil fuel combustion are at a minimum. In order to obtain enough sample for analytical determination, four-hour samples were collected. The logistics of cleaning and preparing the sampling heads made it impractical to shorten the interval between the samples.

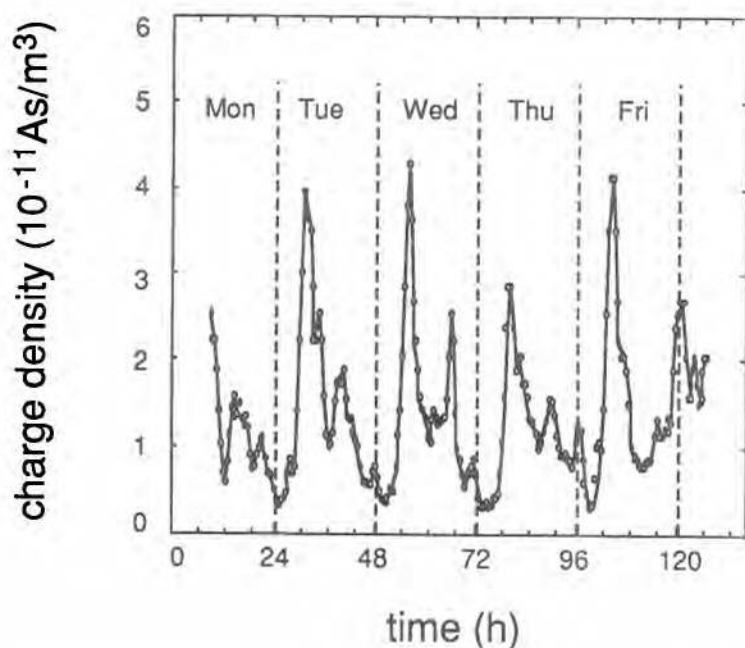


Figure 6. Photoelectric charge density vs. time between May 27 and June 1, 1991 in Zürich, Switzerland (hourly averages).

Figure 7 presents the photoelectric charge densities as a function of PPAH concentrations for all samples. The regression parameters are given in Table 1. A linear correlation ($r^2 = 0.87$) was found between PPAH concentrations and photoelectric charge densities. These results are very encouraging in light of the fact that the samples were collected over a three-week period with a wide variety of environmental conditions. As mentioned in the study with garage TSP (McDow et al., 1990), a number of factors may influence the correlation. The major factor in the lower correlation compared to the oil stove results from McDow et al. (1990) is most likely how long the TSP has been in the atmosphere or the degree of aging that the TSP has undergone between sampling sites and times. The aging of TSP has been examined by various authors and the resulting TSP is a result

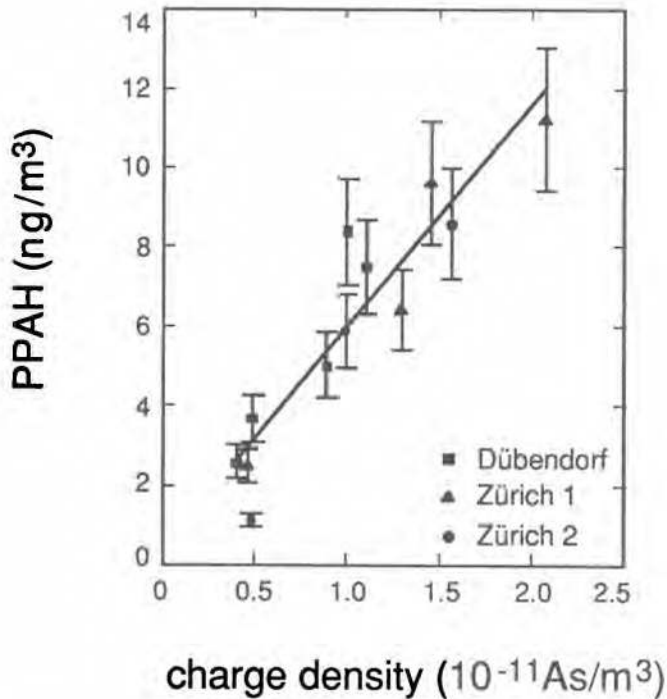


Figure 7. Photoelectric charge density vs. PPAH concentration for Dübendorf and Zürich. The regression parameters are summarized in Table 1. Error bars shown are ± 1 standard deviation.

of a combination of condensation and volatilization of water (Thibodeaux et al., 1991) and volatile and semi-volatile organic compounds (Pankow, 1987, 1988), chemical reactions on the particulate surface (Finlayson-Pitts and Pitts, 1986), and atmospheric removal mechanisms (Bidleman, 1988).

Instead of looking at 12 samples collected over a three week period at two different locations, the data for each sampling period were also examined separately. Figure 7 and Table 1 summarize the correlations between PPAH concentration on TSP and photoelectric charge densities for the three periods. Two points can be made about the data in Figure 7: 1) for each of the three periods, linear correlations ($0.83 \leq r^2 \leq 0.94$) were obtained between the PPAH concentrations associated with TSP and the photoelectric charge densities; and 2) the average ratio of PPAH concentration in Dübendorf to Zürich (1 & 2) was $\sim 75\%$, while the average ratio of photoelectric charge density was 60%. These results suggest that at least two distinct types of TSP were sampled. It can be seen from Table 1 and Figure 7 that there is a slight decrease in the correlation ($r^2 = 0.92$ to 0.87) that is thought to be the result of the addition of the slightly aged TSP data from Dübendorf to the fresh TSP data from Zürich. The results from the Dübendorf regressions indicate that the TSP may be aged aerosols, having a surface coating (e.g. water, sulfate, n-alkanes) that may inhibit APE. The Zürich regressions indicate fresh TSP with relatively strong APE signals and the Dübendorf data indicate TSP that is most likely a combination of fresh TSP from local sources and aging TSP via transport from Zürich. Thus, for the conditions encountered in this field study, the APE technique appears to be well suited as an in-situ, real-time surrogate technique for PPAH determination.

The results are summarized in Figure 8. This plot presents log photoelectric charge density vs. log PPAH concentration data from the oil stove and parking garage (McDow et al., 1990). This shows that for a dynamic range of 3 orders of magnitude and for different kinds of aerosols a reasonable relation between the reading of a photemission sensor and the total particulate PAH concentration exists.

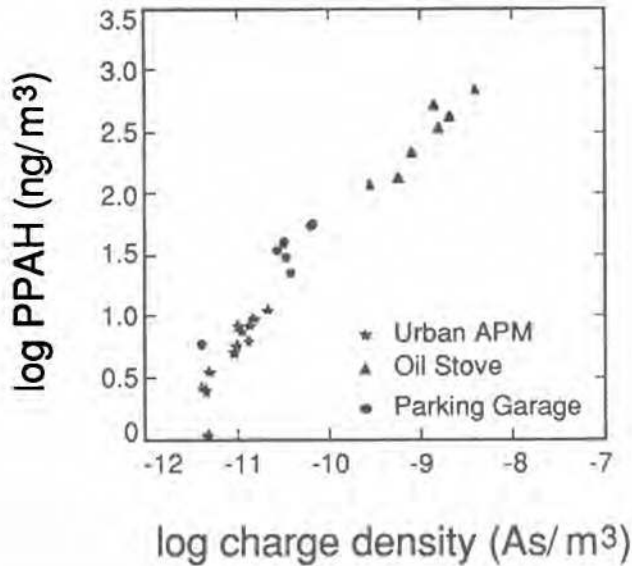


Figure 8. Photoelectric charge density vs. PPAH concentration measured at an oil stove and a parking garage in Hönnggerberg, Switzerland (McDow et al., 1990), and Zürich, Switzerland. The regression parameters are summarized in Table 1. Adapted from Burtscher et al., (1992).

4. Summary and Conclusion

The results obtained from both laboratory and field studies indicate that photoemission is a useful screening technique to monitor PPAH concentrations. The correlations developed can be used to predict PPAH concentrations, which are difficult to obtain experimentally, from easily obtainable photoelectric charge densities. It has been shown that these correlations decrease with increased atmospheric residence time of the particulate matter. This is believed to be a direct consequence of aging of the TSP. Since APE apparatus is relatively inexpensive, there is the potential of using this technique in tracking air parcel trajectories in urban areas. Since PAHs associated with TSP have been shown to be powerful carcinogens and mutagens, the APE technique may be useful as a screening device to examine different fuels and types of internal combustion engines as sources of PPAH emissions. The latter may be implemented in a similar manner to carbon monoxide measurements during vehicle inspections. However, in order to fully understand the mechanisms associated with APE and develop its potential, the cooperation of at least two different disciplines, physicists and

environmental chemists, is required. It is only through projects like WaBoLu that interdisciplinary work of this nature can be completed.

Acknowledgements

The authors would like to thank H. C. Siegmann for his active support of this project and many fruitful discussions, Christian Schaffner from EAWAG for providing technical assistance and Eva Molnar from EAWAG for her helpful comments and assistance in manuscript preparation. Funding by the Swiss Federal Institute of Technology Zürich under the project WaBoLu (Wasser-Boden-Luft, Water-soil-air) is greatly appreciated.

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