

**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

A two year's source apportionment study of wood burning and traffic aerosols for urban and rural sites in Switzerland

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Received: 7 October 2010 – Accepted: 12 November 2010 – Published: 24 November 2010

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

The contributions of fossil fuel (FF) and wood burning (WB) emissions to black carbon (BC) have been investigated in the past by analysis of multi-wavelength aethalometer data. This approach utilize the stronger light absorption of WB aerosols in the near ultraviolet compared to the light absorption of aerosols from FF combustion.

Here we present two years of seven-wavelength aethalometer data from one urban and two rural background sites in Switzerland measured from 2008–2010. The contribution of WB and FF to BC was directly determined from the absorption coefficients of FF and WB aerosols which were calculated by using confirmed absorption exponents and aerosol light absorption cross-sections that were determined for all sites. Reasonable separation of total BC into contributions from FF and WB was achieved for all sites and seasons. The obtained WB contributions to BC are well correlated with measured concentrations of levoglucosan and potassium while FF contributions to BC correlate nicely with NO_x . These findings support our approach and show that the applied source apportionment of BC is well applicable for long-term data sets.

During winter, we found that BC from WB contributes on average 24–29% to total BC at the considered measurement sites. This is a noticeable high fraction as the contribution of wood burning to the total final energy consumption is in Switzerland less than 4%.

1 Introduction

Atmospheric aerosol particles affect chemical, microphysical, and radiative atmospheric processes. They are important when considering both the natural and the anthropogenic climate forcing (Forster et al., 2007). An abundant constituent in atmospheric aerosols is carbonaceous matter (CM), which is composed of black carbon (BC) and organic carbon (OC). BC is the light absorbing part of carbonaceous

AMTD

3, 5313–5342, 2010

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

material, commonly referred to as soot. Compared to other aerosol constituents BC has very different optical and radiative properties, contributing significantly to current global warming (Jacobson, 2001; Forster et al., 2007; Ramanathan and Carmichael, 2008; Jacobson, 2010).

Besides their effects on the Earth's radiation budget, fine carbonaceous particles have been found to cause serious health effects as they penetrate into the human respiratory system (Oberdorster et al., 2002; Jerrett, et al., 2005; Kennedy, 2007). Epidemiologic studies associated BC in particular with respiratory health effects in children and with cardiovascular diseases (Peters et al., 2000; Gauderman et al., 2004).

The major emission sources of soot particles in Europe and Switzerland are diesel engines and incomplete biomass burning. Especially in winter, wood combustion from domestic heating has been found to be a major contributor to air pollution in residential areas (Szidat et al., 2007; Lanz et al., 2008). Given that wood burning (WB) is a CO₂ neutral energy source its impact on air quality is likely to become more and more relevant in the coming years.

Several commercial instruments are available for the determination of BC in particulate matter (PM). In this study, the aethalometer (AE, Hansen et al., 1984) was used for continuous measurement of BC. The working principle of an AE is the following: aerosols are collected on a quartz fibre filter. Subsequently the attenuation of an illuminating light beam is determined. The aerosol absorption coefficient b_{abs} results after correcting the measured light attenuation for artefacts as multiple scattering, “shadowing” and filter-loading (Weingartner et al., 2003; Collaud Coen et al., 2010 and references therein). The BC mass concentration is obtained from b_{abs} divided by the mass specific aerosol light absorption cross section σ_{abs} . Newer AE instruments operate at several wavelengths ranging from the near-ultraviolet (UV) to the near-infrared (IR). The wavelength dependence of the aerosol absorption coefficient b_{abs} can be described by the power law $b_{\text{abs}}(\lambda) \sim \lambda^{-\alpha}$, where λ is the wavelength of the light beam and α is the Ångström exponent. The spectral dependence allows distinguishing

**Source
apportionment of
wood burning and
traffic aerosols**H. Herich et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

carbonaceous aerosols from different sources. This is because of light absorbing OC which in contrast to BC exhibits a stronger absorption at shorter wavelengths (Andreae and Gelencser, 2006; Lukacs et al., 2007; Moosmüller et al., 2009). For example, biomass burning aerosols are known to contain a significant number of light absorbing organic substances or brown-carbon and have a strong spectral dependence ($\alpha > 2$) while emissions from diesel engines contain primarily BC and have a weak spectral dependence ($\alpha \sim 1$) (e.g. Kirchstetter et al., 2004; Clarke et al., 2007). Under certain conditions the range of Ångström exponents for brown carbon and BC may possibly be wider (Lack and Cappa, 2010).

In the past multiple wavelength AEs were deployed to determine the contributions of traffic and WB to total CM. This was accomplished with a source apportionment model introduced by Sandradewi et al. (2008a). Sandradewi et al. (2008a) collected AE data in winter during a measurement campaign in an alpine valley where WB from domestic heating and traffic emissions were the dominating sources of CM. Linear regression of CM against the light absorption coefficient of FF combustion aerosols in the infrared (950 nm) and the light absorption coefficient of WB aerosols in the UV (470 nm) was proposed for source apportionment. The authors estimated an average contribution of WB to total CM of 88%. In a recent study, Favez et al. (2009) applied the same approach to data sampled in urban Paris during a winter field campaign. The authors determined regression coefficients similar to Sandradewi et al. (2008a) and estimated the average contribution of WB to total CM to be 46%.

In this study we applied the AE model to data collected from 2008 to 2010 at three measurement sites in Switzerland. To our knowledge this is the first long-term source apportionment study using this modelling approach. In a first step we focused on CM. Sensitivity tests for different regression models and for various light absorption exponents were performed. It was found that the regression modelling approach is not suitable for long-term datasets because of significant fractions of CM resulting from sources and processes other than FF and WB. Thus in a second step we focused on the contributions of FF combustion and WB to BC which was calculated directly

**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



from b_{abs} by the use of site specific σ_{abs} values. We determined the fractions of BC resulting from emissions of FF combustion and WB on a seasonal and a daily basis and compared our findings with measured concentrations of tracers for WB (levoglucosan and potassium) and with estimated elemental carbon emitted by WB as obtained from a receptor modelling study.

2 Experimental procedure

2.1 Sampling sites and instrumentation

Measurements were performed at one urban and two rural stations of the Swiss National Air Pollution Monitoring Network (NABEL) (EMPA, 2010) from 2008 to 2010. The NABEL station Zurich-Kaserne (ZUE) is an urban background site located in a courtyard in the city centre of Zurich (47°22' N, 8°32' E, 410 m a.s.l.). The location is surrounded by roads with rather low traffic and is not affected by major emissions from industries. The stations Payerne (PAY) and Magadino-Cadenazzo (MAG) are rural-agricultural sites. PAY is located in the western part of the Swiss Plateau one kilometre outside of Payerne, a small city with 8000 inhabitants. The site is surrounded by agricultural land (grassland and crops), forests and small villages (46°48' N, 6°56' E, 489 m a.s.l.). The MAG site is located south of the Alps in the Magadino plane close to the Lago Maggiore (46°09' N, 8°56' E, 204 m a.s.l.) and about two kilometres outside of Cadenazzo, a village with 2000 inhabitants.

Multiple-wavelength AEs (Magee Scientific, USA, model AE31) were deployed at all measurement sites for determination of BC. The instrument continuously detects the aerosol light absorption coefficient at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) with a time resolution of 5 min. Data correction of $b_{\text{abs}}(\lambda)$ was done according to the procedure by Weingartner et al. (2003). All AEs were equipped with $\text{PM}_{2.5}$ inlets.

**Source
apportionment of
wood burning and
traffic aerosols**H. Herich et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Parallel to the measurement of $b_{\text{abs}}(\lambda)$ at the three sites, daily $\text{PM}_{2.5}$ and at MAG daily PM_{10} samples were collected at every twelfth day on quartz fibre filters (Pallflex Tissuquartz 2500QAT) using a high-volume sampler (Digitel DHA-80, $30 \text{ m}^3 \text{ h}^{-1}$ flow rate). Punches of these $\text{PM}_{2.5}$ and PM_{10} filter samples were analysed for organic and elemental carbon (OC and EC) by applying the thermal optical transmission method (TOT). The OCEC analyzer (Sunset Laboratory Inc.) was operated with the EUSAAR2 temperature protocol (Cavalli et al., 2010).

The BC mass concentration is obtained from $b_{\text{abs}}(\lambda)$ divided by the wavelength dependent aerosol light absorption cross section σ_{abs} , denoted as $\sigma_{\text{abs}}(\lambda)$. Here we determined site specific $\sigma_{\text{abs}}(\lambda)$ from the slope of the linear regression of the daily means of $b_{\text{abs}}(\lambda)$ against the EC concentration. Table 2 summarizes the determined site specific values for $\sigma_{\text{abs}}(\lambda)$.

At PAY, OC and EC concentrations are additionally available as 3-hourly mean values from a semi-continuous OCEC analyzer (Sunset Laboratory Inc.; thermal optical transmission method, EUSAAR2 temperature protocol). This instrument was also equipped with a $\text{PM}_{2.5}$ inlet.

Finally, measurements of potassium and levoglucosan in PM_{10} are available from PAY, potassium concentrations are also available in PM_{10} from MAG. Potassium concentrations were determined in daily PM_{10} samples collected at every fourth day from August 2008 to July 2009. Levoglucosan was determined in eight approximately weekly PM_{10} samples collected with a low volume sampler (Rupprecht and Patashnik, model Partisol FRM2000) in fall 2008 and spring 2009.

2.2 The aethalometer model

The AE model aims to quantify the contribution of fossil fuel (FF) and WB aerosol to the BC concentration. The model has been described in detail by Sandradewi et al. (2008a). Briefly, it relies on two assumptions (a) that during winter FF combustion and WB emissions from domestic heating are the dominating sources of CM, and (b) that total ambient CM can be modelled by the light absorption of aerosols emitted by

these two sources. The first assumption implies that the aerosol absorption coefficient $b_{\text{abs}}(\lambda)$ at a given wavelength λ can be expressed as the sum of the light absorption of aerosols emitted by these two sources:

$$b_{\text{abs}}(\lambda) = b_{\text{absFF}}(\lambda) + b_{\text{absWB}}(\lambda) \quad (1)$$

$b_{\text{absFF}}(\lambda)$ and $b_{\text{absWB}}(\lambda)$ are the wavelength dependent aerosol absorption coefficient of BC from fossil fuel and wood burning emissions, respectively. Both quantities can be calculated from light absorption measurements if the spectral dependence (expressed by the absorption exponent α) for both sources are known. With given α_{FF} and α_{WB} and two different wavelength λ_1 and λ_2 , the following equations apply:

$$b_{\text{absFF}}(\lambda_1)/b_{\text{absFF}}(\lambda_2) = (\lambda_1/\lambda_2)^{-\alpha_{\text{FF}}} \quad (2)$$

$$b_{\text{absWB}}(\lambda_1)/b_{\text{absWB}}(\lambda_2) = (\lambda_1/\lambda_2)^{-\alpha_{\text{WB}}} \quad (3)$$

Light absorption measurements at $\lambda_1 = 470$ nm and $\lambda_2 = 880$ nm (or $\lambda_2 = 950$ nm) are used in this approach. This is due to the fact that BC from FF combustion has a weak dependence on wavelength whereas BC from WB shows enhanced absorption at shorter wavelength.

Equations (1)–(3) can be used to calculate $b_{\text{absFF}}(880 \text{ nm})$ and $b_{\text{absWB}}(470 \text{ nm})$. In the approach by Sandradewi et al. (2008a), the mass concentration of total carbonaceous matter was regressed against $b_{\text{absFF}}(880 \text{ nm})$ and $b_{\text{absWB}}(470 \text{ nm})$ for determination of the contribution of fossil fuel combustion and wood burning to CM,

$$\text{CM} = C_1 \cdot b_{\text{absFF}}(880 \text{ nm}) + C_2 \cdot b_{\text{absWB}}(470 \text{ nm}) \quad (4)$$

with the parameters C_1 and C_2 relating the aerosol light absorption coefficient to the total carbonaceous mass concentration.

**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



In this study, $b_{\text{absFF}}(880 \text{ nm})$ and $b_{\text{absWB}}(470 \text{ nm})$ are also directly used to calculate the contributions of FF and WB to total BC (BC_{FF} and BC_{WB}) by using the average site specific $\sigma_{\text{abs}}(\lambda)$ as indicated in Table 2:

$$\text{BC}_{\text{FF}} = b_{\text{absFF}}(880 \text{ nm})/\sigma_{\text{abs}}(880 \text{ nm}) \quad \text{and} \quad \text{BC}_{\text{WB}} = b_{\text{absWB}}(470 \text{ nm})/\sigma_{\text{abs}}(470 \text{ nm}) \quad (5)$$

5 Note that the derived BC_{FF} and BC_{WB} are consistent with elemental carbon concentrations because the aerosol light absorption cross sections were calculated from EC analysis using the thermal optical transmission method.

3 Results

3.1 Light absorption measurements

10 For PAY, MAG and ZUE the absorption exponent α was calculated over all seven wavelengths and for different time intervals. Figure 1 shows time series for the monthly mean values of α from March 2008 to March 2010. A strong seasonal cycle of α can be observed at all sites, the Ångström exponent is higher during winter than during summer. During winter α is similar at the two rural sites PAY and MAG, mean values
15 vary between 1.20 and 1.35. In summer MAG shows lower α values ($\sim 0.9 \pm 0.05$) than PAY ($\sim 1.0 \pm 0.05$). At both stations the observed α values were lower in 2009 compared to 2008. For the urban background site ZUE the monthly α varies from 1 to 1.05 during summer, α is about 1.15 during winter.

20 Figure 2a and b shows average diurnal cycles of α for summer (June–August) and for winter (December–February), respectively. In both figures, α is shown as hourly values calculated from measurements that were taken on working days only. PAY and MAG show similar diurnal patterns during winter, where α varies between 1.35 during night and 1.25 during daytime. In winter α values close to 1.25 are observed during night at ZUE, during day α is less than 1.1 in the morning and close to 1.05 during the
25 evening rush hours. At PAY α varies between about 1.0 (night) and about 0.9 (day).

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Lowest α values of 0.87 are observed during the morning rush hours. At MAG α varies from 0.8 to 0.9 during summer, lowest α values of approx. 0.8 are observed in the morning and evening rush hours. In ZUE, α varies between approx. 1.1 (night) and approx. 1.0 (day) in summer and a daily minimum with α approx. 0.95 occurs during morning rush hours.

The mean α values observed during winter are approx. 1.3 in PAY and MAG and approx. 1.15 in ZUE. These values are lower than what has been reported for biomass burning aerosols (e.g. Kirchstetter et al., 2004; Bergstrom et al., 2007) but very similar to values observed at populated and polluted sites during winter (Favez et al., 2009; Yang et al., 2009). The diurnal patterns show at all stations lowest α during daytime and especially during the morning rush hours. This indicates the influence of particulate matter emitted by road traffic.

Figure 2a and b includes OC/EC ratios derived from 3-hourly concentration measurements at PAY. The mean OC/EC ratio at PAY varies from 4 to 8 during summer with the daily minimum between 06:00 a.m. and 09:00 a.m. and the maximum from 12:00 p.m. to 03:00 p.m. During winter the variability of the OC/EC ratio is reduced, the observed OC/EC ratios vary between 4 and 6.

During summer, Ångström exponents less than 1 are observed at all measurement sites. Such low α values have been reported before (Gyawali et al., 2009 and references therein). Gyawali et al. (2009) show that values of $\alpha < 1$ may occur for large aerosol particles and/or aerosol particles of certain shape and mixing state. For example, the authors attributed $\alpha < 1$ to aerosol particles that consist of a collapsed soot core and a coating shell of organic and inorganic secondary aerosol. Especially in MAG where the lowest α values were observed during summer it is known from filter analyses that organic matter contributes on annual average 40% to the total PM₁₀ mass (M. Gianini, personal communication, 2010). This is high compared to values in rural sites north of the alps.

At PAY, MAG and ZUE α values are in summer approx. 0.9 ± 0.1 during the morning rush hours. At the same time the OC/EC ratio at PAY is at a daily minimum. This

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



observations point to an impact of road traffic emissions, we therefore attribute $\alpha \sim 0.9$ to BC from road traffic emissions. As discussed earlier, previous studies often reported values of $\alpha \sim 1$ for diesel soot experiments. However, site specific traffic values may differ from the laboratory results. Our measurement stations are neither located nearby roadways nor directly situated close to other primary emission sources. Sampled traffic related particulate matter may have likely undergone some aging and may be collapsed or coated by the time the sampling site is reached. Here we choose the absorption coefficient $\alpha_{FF} = 0.9$. The absorption coefficients $\alpha_{WB} = 1.9$ is taken from literature (Sandradewi, 2008a).

3.2 Application of the aethalometer model

3.2.1 Adaptability testing

Sandradewi et al. (2008a) estimated the contribution of FF and WB to carbonaceous matter by multiple linear regression according to Eq. (4). This approach implies that no other sources than FF combustion and WB and no relevant CM formation process (e.g. formation of secondary organic aerosols SOA) are present. This assumption might be adequate during winter and for certain locations as shown in Sandradewi et al. (2008a). The approach was also applied by Favez et al. (2009), both studies found similar model parameters C_1 and C_2 for the short measurement campaigns performed during winter.

In a first try to test this source apportionment approach, we calculated total CM concentration from Eq. (4) by using the values for C_1 and C_2 from (Sandradewi, 2008a; Favez, 2009) and $b_{absFF}(\lambda_1)$ and $b_{absWB}(\lambda_2)$ as derived from our measurements using Eqs. (1)–(3). We found systematic differences between calculated CM and CM determined from measured OC and EC (denoted as measured CM). During summer the calculated CM was less than 50% of the measured CM which might result from large contributions by sources and processes other than FF combustion and WB during summer (e.g. SOA formation). During winter the calculated CM was about 25% larger than measured CM.

In a second try we performed regression modelling (with and without intercept, i.e. C_3) and with varying Ångström exponents α for estimating the contribution of FF combustion and WB to CM (Eq. 4). The modelling was performed for all available AE data as well as for winter data only.

This approach leads to a satisfactory agreement between measured and modelled CM, however, the standard error of the estimated C_1 , C_2 (and where applicable C_3) is around $\pm 30\%$ allowing no meaningful quantification of source contributions. In addition the sensitivity of C_1 and C_2 on the chosen Ångström exponents for aerosols from FF combustion and WB is high leading to a further increase in uncertainty.

Error estimates in the AE model are only sparsely discussed in literature. For example, Sandradewi et al. (2008a) and Favez et al. (2009) give no information about the uncertainty of the estimated parameters of the regression models. Also subsequent studies that include an intercept in the regression approach give only limited information about model errors (Sandradewi et al., 2008b; Favez et al., 2010).

Our investigations imply that the simple two sources approach expressed by Eq. (4) is not adequate for the long-term datasets from the three considered measurement sites because of significant contributions to CM from additional sources and processes.

3.2.2 Source apportionment of BC

There after, we determined the contribution of FF combustion and WB to BC (BC_{FF} and BC_{WB}) at PAY, MAG and ZUE. As described in Sect. 3.1 we used $\alpha_{FF} = 0.9$ and $\alpha_{WB} = 1.9$ to determine b_{absFF} and b_{absWB} . BC_{FF} and BC_{WB} are directly determined from $b_{absFF}(880\text{ nm})$ and $b_{absWB}(470\text{ nm})$ using the site specific values for $\sigma_{abs}(\lambda)$ (see Table 2).

Figure 3 shows the calculated BC_{FF} and BC_{WB} concentrations at the PAY, MAG and ZUE sites from April 2008 to April 2010 as daily mean values. For each measurement site the relationship between total BC and the available EC concentrations in $PM_{2.5}$ collected at every fourth day from August 2008 to July 2009 is also shown in a scatter plot. Table 3 shows seasonal mean concentrations of total BC, and the contributions

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of BC_{FF} and BC_{WB} to total BC for winter and summer at all three stations. The given uncertainty estimates for BC_{FF} and BC_{WB} were determined by varying the associated Ångström exponent α by ± 0.05 . In general, the total BC concentration is in winter at all stations substantially higher than in summer. On the one hand this can be explained by different meteorological conditions, e.g. in winter frequent temperature inversions lead to a reduced vertical mixing of the air and to an accumulation of air pollutants within the boundary layer. On the other hand emissions from WB have an additional impact on total BC concentrations during the cold season.

At the rural measurement site PAY the average BC concentration during winter (DJF mean) is $0.80 \pm 0.07 \mu\text{g m}^{-3}$ with $29 \pm 4\%$ of the total BC resulting from WB emissions. In summer (JJA mean) the BC concentration is on average $0.43 \pm 0.04 \mu\text{g m}^{-3}$ with WB contributions of $9 \pm 3\%$. A similar seasonal pattern for BC can be observed in MAG. This rural site south of the alps shows highest BC concentrations during winter with a mean of $3.03 \pm 0.3 \mu\text{g m}^{-3}$ where $27 \pm 4\%$ of total BC are estimated as BC_{WB} . During summer the mean BC concentration is $0.80 \pm 0.08 \mu\text{g m}^{-3}$, all of the BC during summer is found to result from emissions of FF combustion. The seasonal differences in BC concentrations are much lower at the urban background site ZUE compared to the rural sites. During summer the average BC concentration is $0.99 \pm 0.09 \mu\text{g m}^{-3}$ compared to $1.34 \pm 0.13 \mu\text{g m}^{-3}$ during winter. BC_{WB} accounts for $10 \pm 4\%$ of total BC during summer and $24 \pm 3\%$ during winter. In 2002 and 2003, Szidat et al. (2006) performed ^{14}C analyses of elemental carbon in PM_{10} collected at the measurement site ZUE. The authors reported contributions of biomass burning to EC as $6 \pm 2\%$, $12 \pm 1\%$, and $25 \pm 5\%$ for PM_{10} samples from summer, spring and winter, respectively. These results are in good agreement with the WB contribution determined in this study.

Beside the seasonal averages, the contributions of FF combustion and WB to total BC have also been evaluated on a higher temporal resolution. Diurnal cycles of BC_{FF} , BC_{WB} and total BC have been calculated as 3 h mean values and are shown in Fig. 4.

In summer mean concentrations of BC in PAY are generally below $0.5 \mu\text{g m}^{-3}$, both at weekdays and weekends, all of the BC is identified as BC_{FF} . BC concentrations

**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



are generally lowest during noon and afternoon which can be attributed to an increasing boundary layer height and corresponding aerosol dilution. During weekdays a maximum of total BC concentration occurs during morning rush hours. In winter the total BC concentrations at PAY is $0.8\text{--}1\ \mu\text{g m}^{-3}$ during weekdays with slightly elevated concentrations during rush hours. A BC concentration of approx. $0.5\ \mu\text{g m}^{-3}$ can be attributed to FF combustion during night and at weekends. At weekdays in winter the contribution of BC_{FF} is approx. $1\ \mu\text{g m}^{-3}$ during the morning and evening rush hours. The BC concentration in winter attributed to WB is both at weekdays and weekends relatively constant during the day and increases to $0.4\ \mu\text{g m}^{-3}$ in the evening.

In MAG the diurnal pattern of total BC is in summer similar to the BC pattern in PAY. Total BC can exclusively be attributed to BC_{FF} . During weekdays BC concentrations dominate during the morning rush hours. In winter, BC concentrations in MAG vary from $2.5\text{--}4.8\ \mu\text{g m}^{-3}$ during weekdays and from $1.4\text{--}3.2\ \mu\text{g m}^{-3}$ at weekends. During weekdays BC_{FF} is approx. $3.5\ \mu\text{g m}^{-3}$ during the late morning and the evening rush hours. At weekends and at night BC_{FF} varies from $1.0\text{--}2.0\ \mu\text{g m}^{-3}$. During weekdays and weekends BC_{WB} is lowest during the day, in the evening concentrations are increasing.

In ZUE the BC concentrations are in summer predominantly resulting from traffic emissions, the contribution of BC_{WB} to total BC is negligible except for the evening hours at the weekends. This fraction can be attributed to local emissions from public fire and barbecue places close to the measurement site. In winter the total BC concentration in ZUE varies from $0.8\text{--}1.3\ \mu\text{g m}^{-3}$ during weekends and from $1.2\text{--}1.7\ \mu\text{g m}^{-3}$ during weekdays where highest concentrations occur during the morning and the evening. BC_{WB} concentrations vary from $0.3\text{--}0.5\ \mu\text{g m}^{-3}$, both at weekdays and weekends where the higher concentrations occur during the evening hours.

In summary, the diurnal cycle of BC_{FF} follows at all sites the expected pattern with highest concentrations during the rush hours when road traffic density is at maximum. On the other hand, the contribution of BC_{WB} is only significant during winter. The diurnal cycles show at all sites increased concentrations during evenings

and nights which can be explained by the typical operating time pattern of domestic heating appliances. The absolute concentrations and the found diurnal cycles of BC_{WB} and BC_{FF} indicate that the applied source apportionment approach gives reasonable results.

5 In the following we try a further verification of our approach by relating BC_{FF} and BC_{WB} to independent tracers for FF combustion and WB, respectively.

3.2.3 Comparison with auxiliary data

For PAY the determined BC for FF combustion BC_{FF} and for WB emissions BC_{WB} are plotted against two markers for WB related aerosols, potassium and levoglucosan.

10 Figure 5a shows the relationship between daily potassium concentration and daily BC_{FF} and BC_{WB} at PAY. Daily BC_{WB} and potassium are positively correlated ($R^2 = 0.74$) while there is a much lower correlation between BC_{FF} and potassium ($R^2 = 0.17$). In Figure 5b the relationship between levoglucosan and BC_{FF} and BC_{WB} is shown for PAY. Each data point represents a mean concentration over roughly a one week period. The calculated BC_{WB} correlates well with levoglucosan, resulting in a high coefficient of determination $R^2 = 0.78$. There is no linear dependence between BC_{FF} and levoglucosan ($R^2 = 0.01$).

15 In addition, BC_{FF} and BC_{WB} were compared to EC source contributions as obtained in a PM_{10} source apportionment study (M. Gianini, personal communication, 2010) using a receptor modelling approach (positive matrix factorization PMF, Paatero and Tapper 1994). Figure 5c shows BC_{FF} and BC_{WB} versus the daily contribution of “wood combustion EC” for PAY as obtained by PMF. The correlation between BC_{WB} and “wood combustion EC” is good resulting in a coefficient of determination $R^2 = 0.75$ while there is only little correlation between BC_{FF} and “wood combustion EC” derived by PMF.

25 Also for MAG BC_{FF} and BC_{WB} were plotted against potassium (Fig. 6a). Daily BC_{WB} and potassium are positively correlated ($R^2 = 0.69$). The correlation between BC_{FF} and potassium is clearly lower ($R^2 = 0.49$). In Fig. 6b, BC_{FF} and BC_{WB} were compared

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

to EC source contributions from PMF for MAG. Here, the correlation between BC_{WB} and “wood combustion EC” results in a coefficient of determination $R^2 = 0.74$. There is a clearly lower correlation between BC_{FF} and “wood combustion” derived from PMF ($R^2 = 0.33$).

Note that for PAY and MAG, the findings for potassium and the modelled contribution of “wood combustion EC” are based on long-term measurements, the used data were collected during a whole year.

For the time period of the ZUE measurements neither potassium concentrations nor PM_{10} source apportionment results are available. However, in contrast to the rural sites PAY and MAG, the urban background site ZUE shows typically a high correlation between BC and nitrogen oxide NO_x ($NO_x = NO + NO_2$). Figure 7a and b shows the relationship between daily BC_{FF} and BC_{WB} and NO_x data (provided by NABEL) at ZUE for summer and winter, respectively. For both seasons the correlation between BC_{FF} and NO_x is good resulting in a high coefficient of determination $R^2 = 0.52$ (summer) and $R^2 = 0.73$ (winter). There is no correlation between BC_{WB} and NO_x .

The comparisons show that there is in general a good agreement between the derived BC_{WB} and BC_{FF} with concentrations of WB and FF combustion indicators. These findings give confidence that the applied source apportionment approach for BC is well suited for long-term data sets.

Similar to the above described comparisons, Sandradewi et al. (2008b) found rather high correlations between the optical absorption of aerosols from FF combustion and WB and the fractions of EC that are of fossil and non-fossil origin. The latter was derived from analysis of ^{14}C in EC. The results from Sandradewi et al. (2008b) are therefore also indicating the high potential of data from multi-wavelength aethalometers for identification of the fractions of BC from FF combustion and WB.

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



4 Conclusions

The AE is a robust and easy to use instrument for continuous optical determination of BC concentrations with temporal resolution of a few minutes. In this study, the measured BC concentrations are consistent with elemental carbon concentrations because the aerosol light absorption cross sections were calculated from EC analyses using the thermal optical transmission method.

We deployed multi-wavelength AE instruments at two rural (PAY and MAG) sites and one urban background site in Switzerland. The measurements were performed for up to two years. We found average black carbon concentrations of $0.43 \mu\text{g m}^{-3}$ at PAY, $0.8 \mu\text{g m}^{-3}$ at MAG and $0.99 \mu\text{g m}^{-3}$ at ZUE in summer and $0.8 \mu\text{g m}^{-3}$ at PAY, $3.03 \mu\text{g m}^{-3}$ at MAG and $1.34 \mu\text{g m}^{-3}$ at ZUE in winter.

Recent studies give reason that AE data may be used for source apportionment (Sandradewi et al., 2008a; Favez et al., 2009). But in these studies, the contribution of fossil fuel combustion and wood burning to the total carbonaceous aerosol was determined by analysis of AE data predominantly collected in winter during short-term measurement campaigns. Here we showed that the proposed modelling approach is not applicable for long term datasets. This is due to significant fractions of the carbonaceous aerosol resulting from other sources and processes than FF combustion and WB.

In this study we focused on source apportionment of BC instead of total carbonaceous matter. The modified two sources approach fits very well to the measured BC concentrations. Separation of total BC into BC_{FF} and BC_{WB} was successful for all seasons and measurement sites. In winter, the determined mean fraction of BC_{WB} to total BC was 29%, 27% and 24% at PAY, MAG and ZUE respectively. These results are noticeable with respect to air quality control as wood combustion only contributed 3.9% to the total energy consumption in 2008 in Switzerland (Kaufmann, 2009).

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



But it is interesting to note that the calculated contribution of BC_{WB} is in excellent agreement with results reported for ZUE based on ^{14}C analyses (Szidat et al., 2006). Also, the obtained WB contributions to BC at PAY and MAG correlated well with measured concentrations of levoglucosan and potassium as well as with results from PM_{10} factor analytical modelling. In ZUE there is a good correlation between the obtained BC from FF combustion and NO_x . The latter findings support our approach and show that multi-wavelength AE data are suitable for source apportionment of BC.

Acknowledgements. This study represents part of the research project IMBALANCE funded by the Competence Center Environment and Sustainability of the ETH Domain (CCES). Support from the Swiss Federal Office for the Environment (FOEN) is gratefully acknowledged. Many thanks to André Prévôt and Rudolf Weber for helpful discussions.

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Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Table 1. Measurements performed at the Zurich-Kaserne (ZUE), Payerne (PAY) and Magadino-Cadenazzo (MAG) sites.

Site	AE	OCEC analyzer	High vol. sampler	High vol. sampler	Low vol. sampler
	10 min – $b_{\text{abs}}(\lambda)$ in $\text{PM}_{2.5}$	3 h-OC/EC in $\text{PM}_{2.5}$	24 h-OC/EC in $\text{PM}_{2.5}$ (every 12th day)	Potassium in PM_{10} (every 4th day)	Levoglucosan in PM_{10} (weekly samples)
ZUE	Apr 2009–Mar 2010		Mar 2008–2010		
PAY	Mar 2008–2010	Mar 2009–2010	Mar 2008–2010	Aug 2008–Jul 2009	Sep–Oct 2008, Feb–Mar 2009
MAG	Mar 2008–2010 (Instrument failure Dec 2009–Feb 2010)		Mar 2008–2010 (in PM_{10})	Aug 2008–Jul 2009	

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Table 2. Wavelength-dependent aerosol light absorption cross sections used in this study. Values were determined from the linear regression of $b_{\text{abs}}(\lambda)$ against the concentration of elemental carbon in $\text{PM}_{2.5}$ (for MAG in PM_{10}).

λ	Manufacturer	PAY	MAG	ZUE
$\sigma_{\text{abs}}(\lambda) [\text{m}^2 \text{g}^{-1}]$				
370 nm	30	36.7 (± 3.4)	27.3 (± 2.1)	25.3 (± 4.2)
470 nm	23.6	25.8 (± 2.1)	18.8 (± 1.3)	20.8 (± 3.3)
520 nm	21.3	22.1 (± 1.8)	16.4 (± 1.1)	17.1 (± 2.8)
590 nm	18.8	19.2 (± 1.6)	13.9 (± 1.0)	15.0 (± 2.4)
660 nm	16.8	16.8 (± 1.5)	12.1 (± 0.8)	15.3 (± 2.6)
880 nm	12.6	11.9 (± 1.1)	8.5 (± 0.7)	10.7 (± 1.7)
950 nm	11.7	10.6 (± 1.0)	7.6 (± 0.6)	10.3 (± 1.8)

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[⏪](#)
[⏩](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

Table 3. Contribution of BC_{FF} and BC_{WB} to total BC at the PAY, MAG and ZUE sites. Note that the given absolute BC values are consistent with EC as derived with the thermal-optical transmission method (TOT) using the EUSAAR 2 temperature protocol.

	PAY (rural)		MAG (rural)		ZUE (urban)	
	Summer	Winter	Summer	Winter	Summer	Winter
BC_{FF} (%)	91 ± 3	71 ± 4	100 ± 2	73 ± 4	89 ± 4	76 ± 3
BC_{WB} (%)	9 ± 3	29 ± 4	0 ± 2	27 ± 4	10 ± 4	24 ± 3
Total BC ($\mu\text{g m}^{-3}$)	0.43 ± 0.04	0.80 ± 0.07	0.80 ± 0.08	3.03 ± 0.30	0.99 ± 0.09	1.34 ± 0.13

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

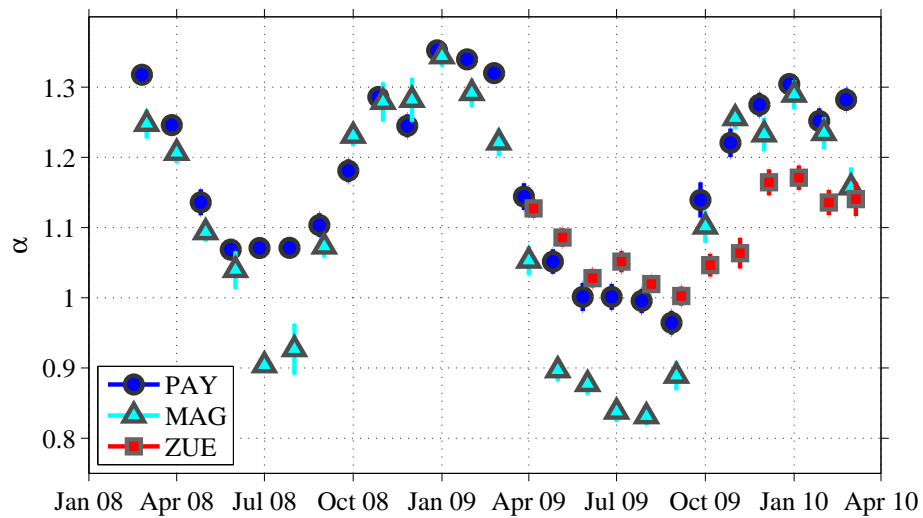


Fig. 1. Time series of the monthly mean Ångström exponent α at PAY, MAG and ZUE. Vertical lines indicate the uncertainty band of the estimated mean values.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

⏪ ⏩

⏴ ⏵

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of wood burning and traffic aerosols

H. Herich et al.

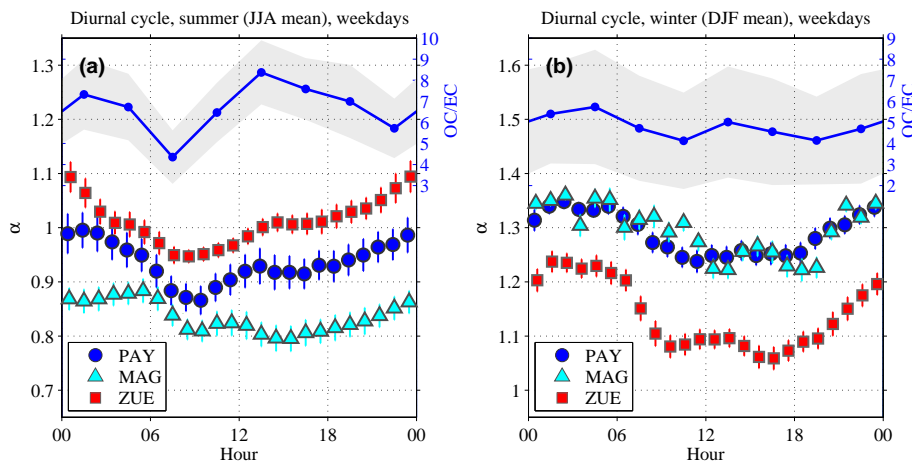


Fig. 2. Average diurnal cycle of hourly α at PAY, MAG and ZUE on weekdays during summer (June, July, August; left panel) and during winter (December, January, February; right panel). Vertical lines indicate the uncertainty band of the estimated mean values. For PAY the OC/EC ratio with one standard deviation band (grey area) is shown (3 hourly means, right axis).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of wood burning and traffic aerosols

H. Herich et al.

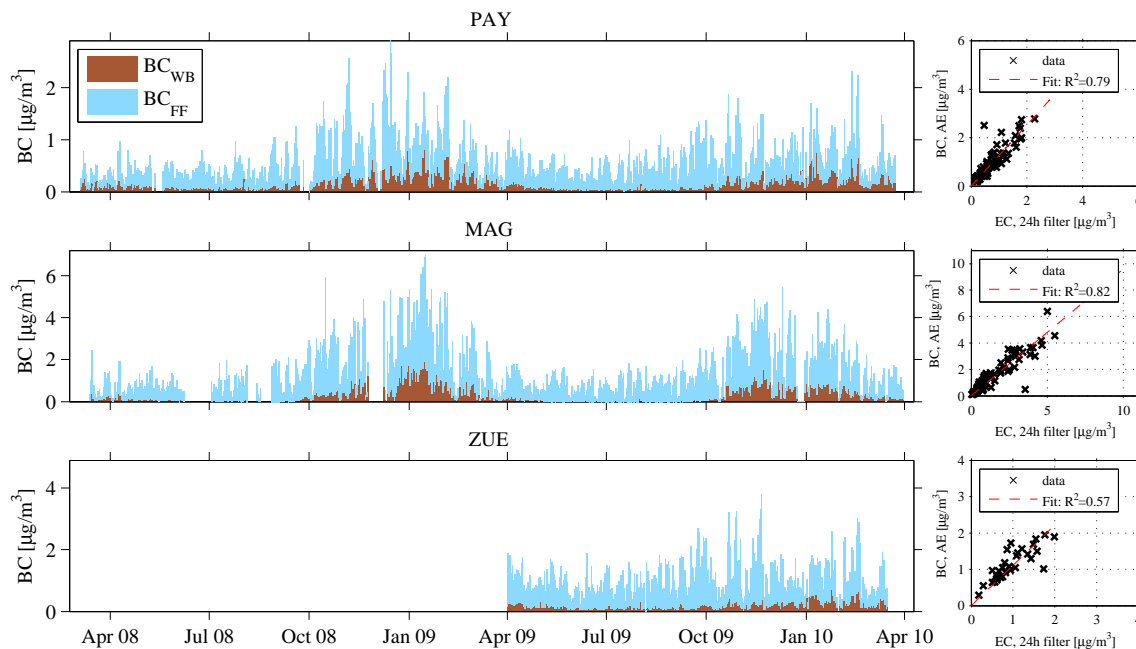


Fig. 3. Long-term pattern of BC_{FF} and BC_{WB} concentrations at PAY, MAG and ZUE (stacked). The scatter plots show the relationship between total daily BC and the daily EC concentration as determined with the thermal optical transmission method at every 4th day from August 2008 to July 2009 for the corresponding measurement site.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of wood burning and traffic aerosols

H. Herich et al.

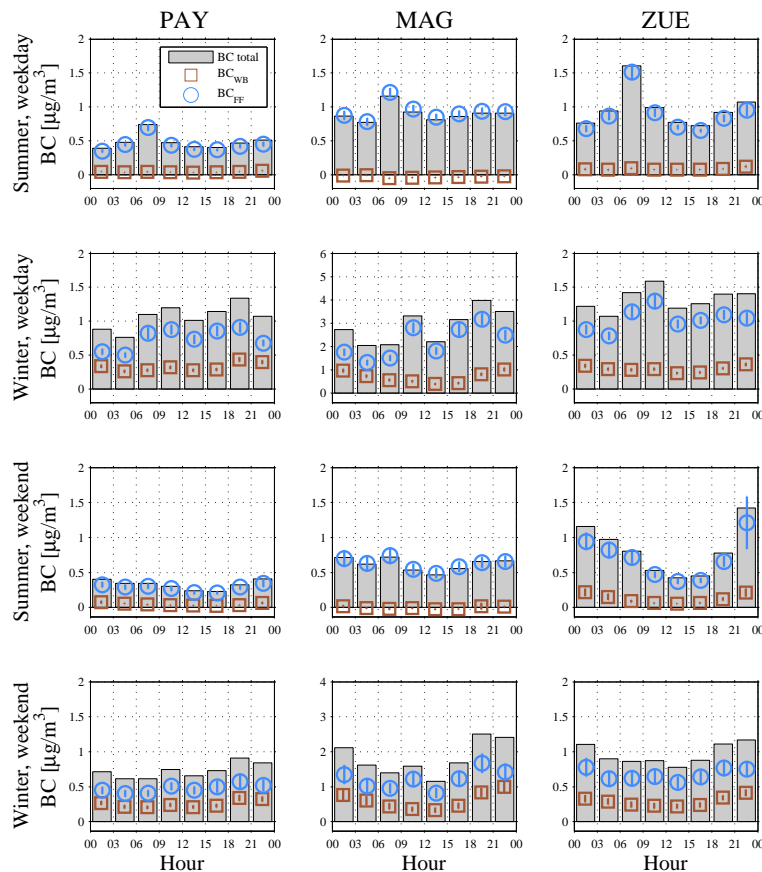


Fig. 4. Diurnal cycles of BC_{FF} , BC_{WB} and total BC concentrations at PAY (column 1), MAG (column 2) and ZUE (column 3). The widths of the uncertainty bands for the estimated mean values are indicated as vertical lines in the symbols. At every site the data were filtered and analyzed for different conditions: weekdays in summer, weekdays in winter, weekend in summer and weekend in winter which are presented in rows 1 to 4, respectively.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of wood burning and traffic aerosols

H. Herich et al.

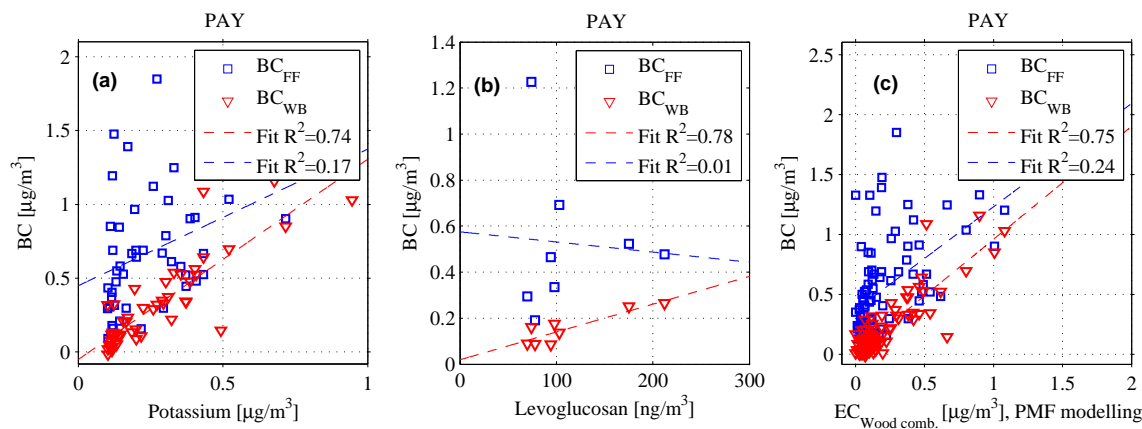


Fig. 5. Scatter plot of daily BC (BC_{FF} and BC_{WB}) and potassium for PAY **(a)**. Scatter plot of daily BC (BC_{FF} and BC_{WB}) and levoglucosan in PM₁₀ for PAY **(b)**. Scatter plot of daily BC (BC_{FF} and BC_{WB}) and “wood combustion aerosol” derived from PM₁₀ factor analytical modelling for PAY **(c)**. All plots include linear regression lines.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source
apportionment of
wood burning and
traffic aerosols

H. Herich et al.

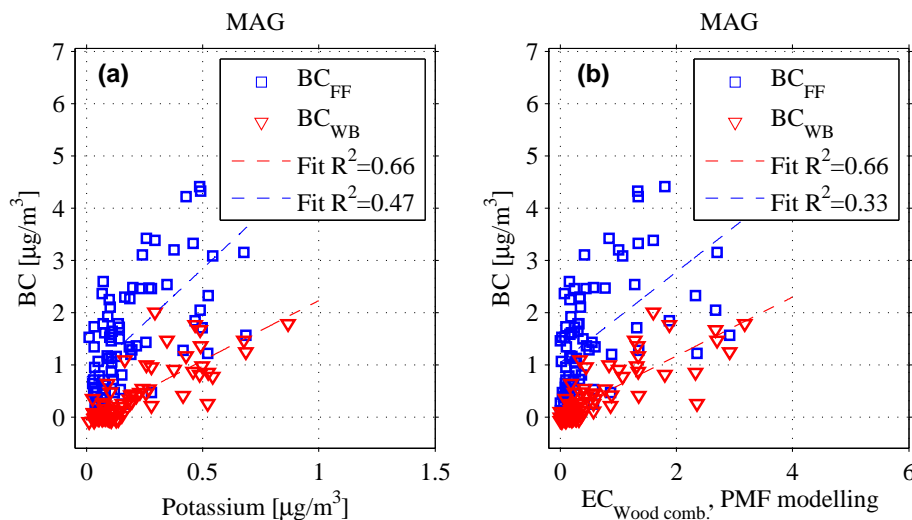


Fig. 6. Scatter plot of daily BC (BC_{FF} and BC_{WB}) and potassium for MAG (a). Scatter plot of daily BC (BC_{FF} and BC_{WB}) and “wood combustion aerosol” derived from PM10 factor analytical modelling for MAG (b). Both plots include linear regression lines.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Source
apportionment of
wood burning and
traffic aerosols**

H. Herich et al.

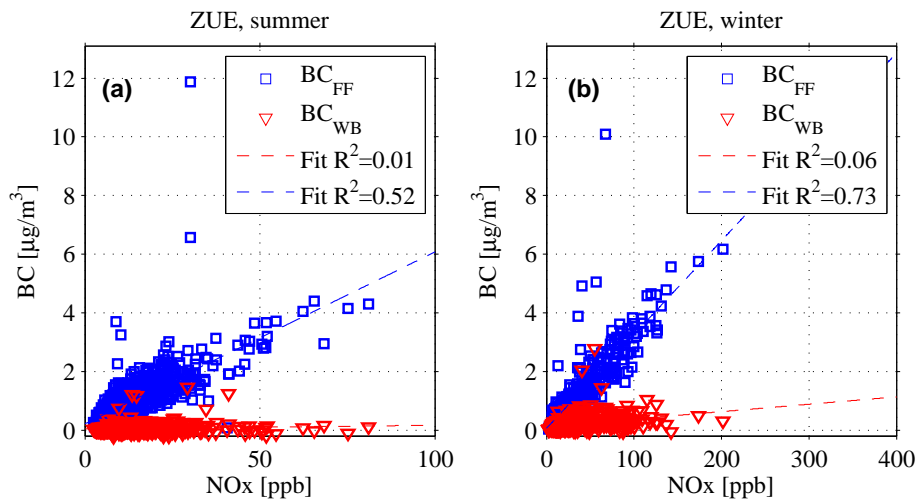


Fig. 7. Scatter plot of daily BC (BC_{FF} and BC_{WB}) and NO_x for ZUE for summer (a) and winter (b). Both plots include linear regression lines.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion