

Black and brown carbon

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1. Introduction

Carbonaceous material plays an important role in atmospheric aerosols. Its origin can be anthropogenic (emissions of primary particles and precursor gases for secondary organic aerosol mainly by combustion processes and emissions of volatile organic carbon by various processes) or biogenic (emission of primary particles and precursor gases by plants or through the degradation of biological material). In this section, the focus is on black carbon (BC) or elemental carbon (EC) and "brown carbon" (BrC), which originates mainly from biomass combustion, although other sources are also possible (e.g. Gelencser et al. 2002). BC is formed by incomplete combustion of carbonaceous (mostly fossil) fuels and emitted directly as primary particles. BC and BrC absorb visible light and therefore play a role in anthropogenic climate change (IPCC 2007) and both have been identified as important contributors to health effects of anthropogenic aerosols (BC: Pope and Dockery 2006; wood smoke: Bolling et al. 2009). As wood smoke contains BrC, a section on wood smoke studies in Austria is also included here.

The nomenclature is rather operational. Organic carbonaceous material is usually called organic carbon (OC). This material does not absorb visible light and is thermally unstable. Depending on the substances, some OC can be highly volatile already at ambient temperatures. BC or EC are strongly light absorbing and are not considered to be organic material. Typically BC or EC consist of >95% carbon and may have some graphitic structure. If analysis of this fraction is performed with optical methods, the term black carbon (BC) is used, because of its strong absorption of all wavelengths in the visible spectrum. If analyses are performed with thermal methods, the term elemental carbon (EC) is used, although "thermally refractory carbon" might be more appropriate. In both thermal and optical techniques, separation of EC and BC from OC is quite difficult. One of the major confounders for both optical and thermal techniques is brown carbon (BrC), a mixture of high-molecular weight organic compounds which absorb visible light to some extent with higher absorption at short wavelengths, and which is thermally quite refractory (Andreae and Gelencser 2006). Wood smoke contains BrC, but also e.g. Na and K, which also interfere with thermal measurements of OC and EC and can have quite some influence of the OC/EC split.

2. Methods to measure BC (or EC) and BrC

The methods can be grouped roughly in two categories: optical and thermal, although the thermal methods also use optical techniques to improve the separation between OC and EC.

In Austria, two teams of scientists located at the Vienna University of Technology (Heidi Bauer) and at the University of Vienna (Regina Hitzemberger) are active in developing methods and in method inter-comparison efforts. At this time, all methods are used by one or both Viennese teams unless indicated otherwise. The team at the University of Vienna specializes more on optical and thermo-optical techniques, while the team at the Vienna University of Technology specializes more on thermal and chemical techniques, although there is quite some overlap between the groups.

Optical methods

Most optical methods measure the transmission (or reflection) of light through (or by) a sample containing BC. The resulting signal for light attenuation has to be converted to a mass concentration of BC, which is usually performed by using specific attenuation coefficients of BC obtained from a combination of the optical method and a thermal one. Optical techniques often suffer from losses of light scattered by the particles, which is erroneously attributed to light absorption resulting in an overestimation of the BC concentration. The integrating sphere technique, which will be described separately, needs to be calibrated with proxy substances for BC, but this calibration is not linked to a thermal method to determine EC.

Currently three optical on-line methods are widely used: the multi-angle absorption photometer (MAAP) developed by Petzold and Schönlinner (2004, single wavelength), the particle soot absorption photometer PSAP (Bond et al. 1999, three wavelengths) and the aethalometer developed by Hansen et al. (1984, currently single-wavelength, seven-wavelength and single-wavelength micro-aethalometer instruments are available). The calibration constant for the aethalometer is only obtained from thermal measurements on Diesel soot, while the other optical methods apply additional correction procedures based on radiative transfer calculations (the MAAP) or other correction schemes (e.g. the aethalometer correction by Weingartner et al. 2003, or the PSAP correction by Virkkula et al. 2005).

Off-line methods include the light transmission method (LTM, based on the original paper by Hansen et al. 1984), the integrating plate method (originally developed by Lin et al. 1973, and later extended also by Horvath 1997), and the integrating sphere technique, which has recently been extended to measure

also BrC and is currently the only method that gives quantitative estimates of BrC from atmospheric samples.

Except for a multi-wavelength aethalometer and a PSAP, all optical methods except the LTM are available at the University of Vienna. The LTM is available at the Vienna University of Technology.

Thermal methods

Thermal methods rely on the different temperature sensitivity of organic and elemental carbon. Volatile and semi-volatile material evolves from the samples when they are heated up from room temperature to temperatures of some hundred °C (specified by the temperature protocols of the different instruments or analysis procedures), while elemental carbon stays stable up to ca. 450°C even in oxidizing conditions. In these methods, the samples are heated in a carrier gas stream and the evolving carbonaceous material is converted either to CO₂ or CH₄, which is analyzed quantitatively. The carrier gas for the evolved material can be inert (usually Helium) or contain oxygen (from small amounts of O₂ added to the inert carrier gas to pure O₂). One of the earliest methods was the thermogram method developed at the Vienna University of Technology (Malissa et al. 1981) which, however, had to be modified because of the thermal behaviour of OC.

Organic material, which normally does not absorb visible light, can blacken ("char") when exposed to elevated temperatures. This pyrolytically formed "elemental" carbon (PEC) is thermally quite stable and would lead to an underestimation of OC and a concurrent overestimation of EC if no correction were applied in the thermal methods. In the now rather historic Cachier method (Cachier et al. 1989), different hold times were chosen for the two-step temperature protocol to minimize charring. Most currently used thermal methods record the signal of a laser beam transmitted through or reflected from a sample and set the split point for the OC/EC separation when the laser signal has reached again its initial (i.e. pre-charring) intensity. Among those thermal-optical techniques are the current thermogram of the Vienna University of Technology (described in detail by Jankowski et al. 2008), the Sunset Analyzer (Birch and Cary 1996, used in the NIOSH-Protocol) and the thermo-optical method developed by Chow et al. (1993, used in the IMPROVE-Protocol). In the recent EUSAAR project, the various temperature protocols and charring correction schemes were analyzed for background aerosols and a new protocol was developed (EUSAAR protocol, Cavalli et al. 2010). As atmospheric samples can also contain carbonates, which typically originate from soils or building materials, carbonate corrections should also be performed, e.g. the correction scheme developed recently at the Vienna University of Technology by Jankowski et al. (2008).

In general, thermal methods depend crucially on the separation procedure to differentiate OC and EC. If only Diesel soot is present in a sample, the separation is rather straight-forward, but wood smoke contributions to the aerosol pose substantial problems. Wood smoke contains thermally refractory organic material, which should be attributed to OC, and trace elements such as Na or K which lower the combustion temperature of EC. In the first case, EC will be overestimated, while in the second it will be underestimated. At this time, the "correct" way to deal with samples influenced by wood smoke in the thermal analyzers is still under debate. In a current study, these interferences are quantified by the University of Vienna team (Hitzenberger and Rosati 2010).

Sunset analyzers capable of performing all temperature protocols as well as charring corrections by transmission and reflection are available at both the University of Vienna and the Vienna University of Technology, where measurements can also be performed with the thermogram and Cachier methods. A semi-continuous Sunset Analyzer (Bae et al. 2004) is available at the Umweltbundesamt.

Other methods

Aerosol light absorption and consequently BC can be measured in situ with a Photoacoustic Absorption Spectrometer (PAS, Arnott et al. 1999) or a cavity-ring down instrument (Moosmüller et al. 2005), although these techniques are quite expensive and are not used at the moment in Austria.

Measurement of black and brown carbon – integrating sphere method

The integrating sphere technique (developed originally by Heintzenberg et al. 1982) was extended by Hitzenberger et al. (1996) to measure BC in atmospheric samples. In this technique, a sample is introduced into an integrating sphere, i.e. a sphere coated inside by a perfectly diffusing surface. The interior of the sphere is illuminated with monochromatic light, and the light flux is measured with a silicon photo diode. In the presence of a light absorbing sample, the light flux is decreased, and this decrease can be linked to e.g. EC using a calibration curve obtained with a suitable EC proxy. As light scattered by the sample is also collected by the detector, the IS method does not suffer from losses of scattered light as do the usual filter-based light transmission methods (e.g. the aethalometer or the PSAP). A detailed description of the operating principle and the advantages of the technique compared to other optical techniques is given by Hitzenberger and Tohno (2001). The need to calibrate the sphere with proxy substances, which is quite a complex procedure, actually led to the development of a quantitative estimation of the amount of BrC concur-

rently with the determination of BC (Wonaschütz et al. 2009). The different wavelength dependence of light absorption by BC and BrC, which is visible in the wavelength dependent calibration curves obtained with proxies for Diesel soot and BrC, is used for the quantitative differentiation between these two parameters. This new extension of the integrating sphere method is up to now the only way to arrive at concentrations of BrC in the atmospheric aerosol.

Determination of biomass smoke

As biomass smoke is probably the largest single source for BrC (Andreae and Gelencser 2006), some discussion should be included in this chapter. Methods to analyze atmospheric samples for biomass smoke as well as for source attribution of OC to biomass smoke are an active research area of the Vienna University of Technology team.

Biomass smoke is a complex mixture of organic and inorganic substances, which depends also on the fuel type and the firing conditions. Particulate emissions from manually fired combustion systems are composed mainly of organic compounds and EC while emissions from automatically fired systems are around an order of magnitude lower and contain mostly inorganic components (Schmidl et al. 2011). As most of the identifiable substances in biomass smoke also have other sources, tracers and tracer profiles are used to determine the amount of wood smoke in atmospheric samples. Traditionally, potassium (K) was used as a tracer for wood smoke (Fine et al. 2001), but recently, anhydrosugars formed during the combustion process have come into use. Levoglucosan, mannosan and galactosan are analyzed by high performance liquid chromatography (HPLC) and used to determine the concentration of biomass smoke. Levoglucosan is a major by-product from cellulose pyrolysis and a marker for wood combustion in ambient samples (Simoneit et al. 1999). In a recent study on smoke samples from common Central European wood types, Schmidl et al. (2008a) found that levoglucosan contributed 4–15% by weight to the smoke. Mannosan, which is formed during pyrolysis of hemicelluloses, was found useful to distinguish between softwood and hardwood combustion, as smoke from conifers contains at least four times higher concentration than smoke from deciduous trees, but with 0.4% the content is much lower than that of levoglucosan. Galactosan concentrations in woodsmoke are 10–50 times lower than levoglucosan concentrations, but about equal in smoke from open burning of leaves (Schmidl et al. 2008b). The ratio of levoglucosan/galactosan may therefore help to identify pollution episodes influenced by leaf burning emissions such as from open garden fires.

3. Measurements of BC and BrC in Austria

Method inter-comparison studies – general

Despite more than three decades of intensive efforts, no standard technique for BC (or EC) measurements exists. Over time, dozens of method inter-comparison studies were performed, but usually only two or a few methods were compared. Watson et al. (2005) give a fairly comprehensive overview over inter-comparison studies conducted up to about 2004.

As the Austrian scientists working in the field of carbonaceous particles in the atmosphere are part of several international networks, important international method inter-comparison studies were coordinated by the teams of the Vienna University of Technology (Schmid et al. 2001) and the University of Vienna (Hitzenberger et al. 2006, Reisinger et al. 2008) or performed in cooperation with these teams (Currie et al. 2002, ten Brink et al. 2004). Regina Hitzenberger is currently a member of the CEN/TC 264 Working Group 35 "EC/OC in PM", which endeavours to set a standard measurement method for EC and OC on PM_{2.5} samples. The Vienna University of Technology group participated in the EU projects CARBOSOL and POLYSOA, and the University of Vienna group in the EUROTRAC-II Field project InterComp2000 (coordinated by K. Müller, IfT Leipzig).

Very comprehensive studies involving a large set of techniques and laboratories were performed by Schmid et al. (2001) on a limited set of filter samples obtained from an urban kerbside site and by Currie et al. (2002) on a PM reference material. A recent large-scale method inter-comparison study was part of the EUROTRAC-II project InterComp2000, where atmospheric aerosols were sampled in an intensive field campaign at the rural site Melpitz, Germany (ten Brink et al. 2004) in spring 2000. In general, the large-scale studies show that total carbon (the sum of OC and EC) can be determined within less than 10% with the different thermal methods employed at different laboratories.

For EC, however, results obtained by different methods can vary by an order of magnitude (Schmid et al. 2001) for kerbside samples if thermal methods without charring correction are included in the data set. If only thermal methods with charring correction are considered, the variation is much lower (ca. 25%) and the EC concentrations are lower by ca. 20% than the average over all methods used in the study. For background (or rural aerosol) in springtime in Melpitz, the picture is similar. Method differences for EC and/or BC were found to be within a factor of three (ten Brink et al. 2004), although the time series of BC or EC were highly correlated.

Method inter-comparison studies in the Vienna aerosol – influence of BrC

In summer 2004, an intensive international inter-comparison study was performed in the roof laboratory of the University of Vienna team (Hitzenger et al. 2006). Practically all methods in use at that time in Europe were run in parallel to obtain BC and EC concentrations. The results were highly surprising, as no significant difference could be found between the methods. Although there was of course quite some variation on a day-by-day basis (Figure 1), the campaign averages obtained from the different methods were within their respective standard deviations.

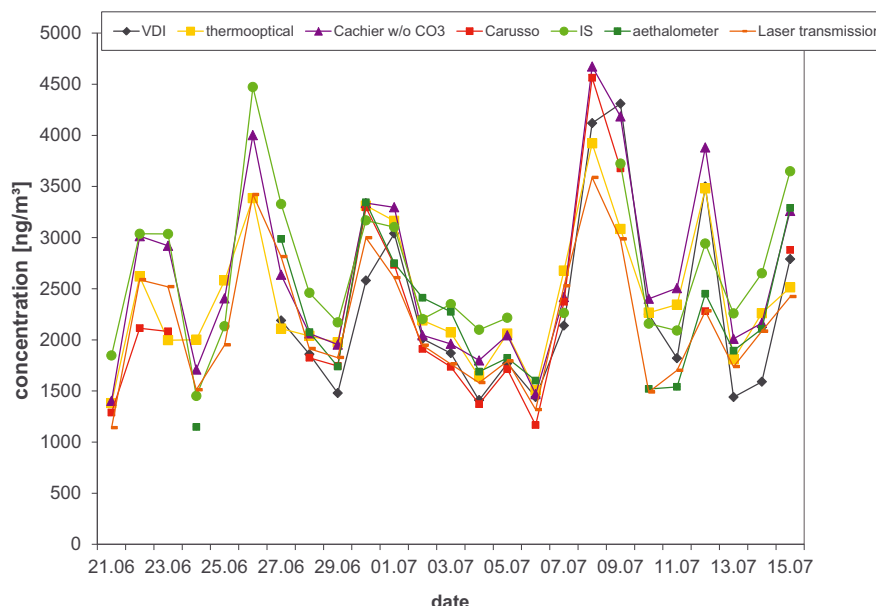


Figure 1: Daily variation of BC and EC during the summer method inter-comparison study in Vienna, all data. (from: Hitzenger et al. 2006). "Carusso" is the old acronym for the MAAP instrument.

In winter 2006, the study was repeated (Reisinger et al. 2008). On mild days, method differences were similar to summer, but on cold days, method differences of up to factors of 4 were found. The highest values were obtained by the Cachier method and the MAAP, and the lowest by the two different thermal protocols (TOT (A3) and TOT (NIOSH) of the Sunset analyzer (Figure 2a). At that time, the method to separate BC and BrC (Wonaschütz et al. 2009) was already available. We found that method differences were most severe when the BrC content of the aerosol was high. The BC concentrations obtained from the integrating sphere after the separation of BrC and BC were in the range of the

lower concentrations, but still appreciably above the concentrations measured with the Sunset instrument (both protocols, Figure 2b).

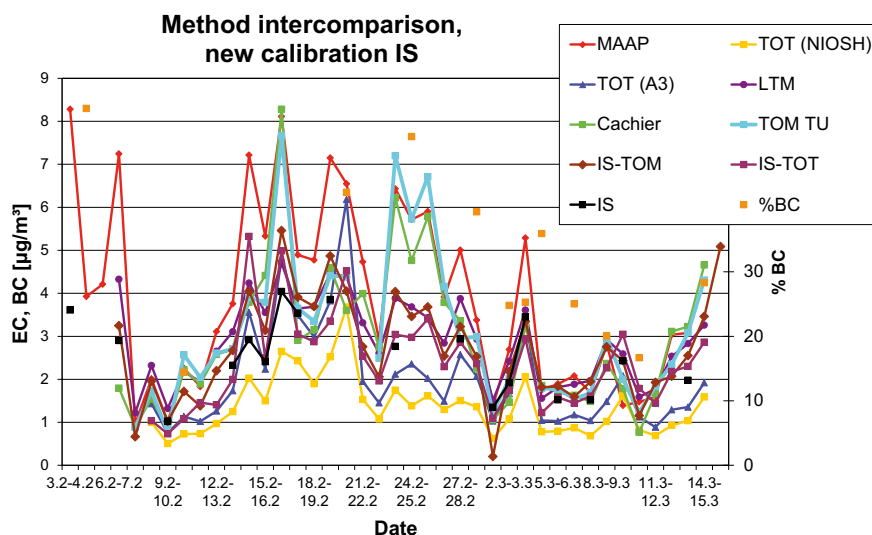


Figure 2a: daily variation of BC and EC during the winter method inter-comparison campaign in Vienna; all data. (from: Reisinger et al. 2008).

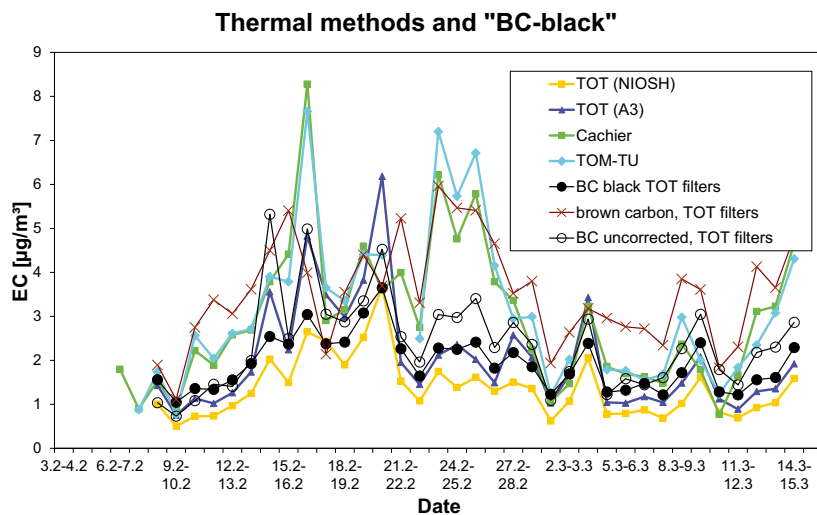


Figure 2b: daily variation of EC and BC (only integrating sphere, corrected and uncorrected for BrC) and BrC during the winter method inter-comparison campaign in Vienna (from: Reisinger et al. 2008).

Brown carbon during a biomass burning episode (Easter fires) in Styria

In rural Austria, garden waste was usually burnt in open fires until air quality regulations prohibited these fires several years ago. Nowadays, fires may be lit only for special occasions, such as the traditional log fires on mid-summer eve and during the Easter night. According to regulations, only dry wood may be burnt, but quite often garden waste collected in the preceding fall is quietly disposed of in the fires. Air pollution can be severe especially in valleys under stable meteorological conditions. One such episode was traced during the measurements for AQUELLA in 2004 (Bauer et al. 2007) in Styria, where PM₁₀ concentrations reached $>100 \mu\text{g}/\text{m}^3$, which was more than twice the 24hr standard value. The results of the EC/BC/BrC measurements were reported by Wonschütz et al. (2009). Figure 3 shows the BC and BrC concentrations obtained with the integrating sphere and EC obtained with the Cachier method and the thermogram TOM-TU method from PM₁₀ filter samples collected in Graz at a kerbside site (Don Bosco), a suburban site (Graz Süd) and a background site (Bockberg) during the Easter weekend 2004. An additional filter sample from Peggau (Easter Sunday 2006) is also included. On Good Friday 2004 (April 9), BrC was present only at one site (Graz Süd) in a very low concentration.

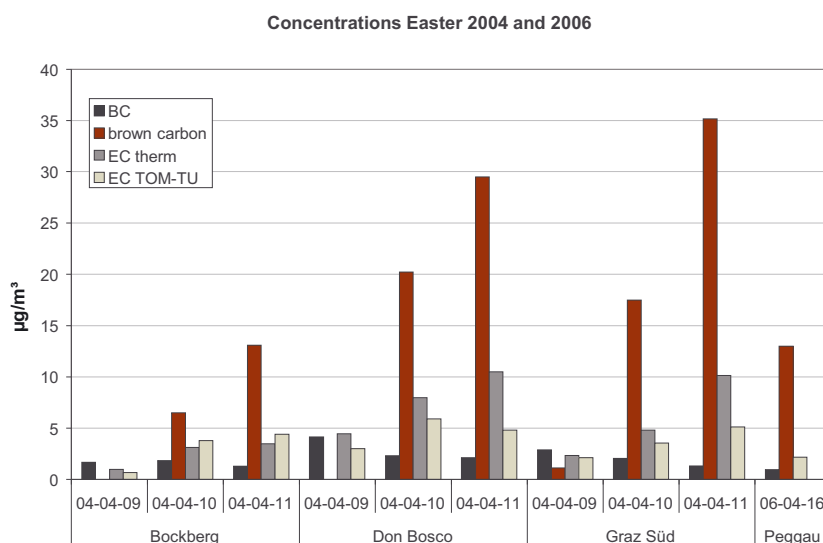


Figure 3: concentrations of BC (traffic derived), BrC (biomass source) and EC determined with the Cachier (EC therm) and thermogram (EC TOM-TU) methods during the Easter weekend 2004 and during Easter Sunday, 2006 (from: Wonschütz et al. 2009).

Fires were lit during the afternoon of Holy Saturday (April 10) and continued burning during the night. BrC concentrations increased drastically at all sites, and method differences became appreciable with largest concentrations measured by the Cachier method within Graz and the thermogram method at the background station. BC, which was corrected for the influence of BrC, follows the expected traffic patterns with highest concentrations on Friday and lowest on Sunday. The other methods overestimated BC quite severely when BrC concentrations were non-negligible during Easter night and Easter Sunday. Similar method differences were found also for the other Easter sample (Peggau 2006).

4. Wood smoke

Wood smoke is studied by the Vienna University of Technology group both in terms of emissions and emission factors and in terms of wood smoke contribution to ambient aerosols.

In a recent study (Caseiro et al. 2009) the impact of wood burning in Austria was investigated for traffic impacted, residential and background sites in Vienna, Graz and Salzburg. At all sites, wood combustion contributed significantly to PM₁₀ especially during the cold season, when wood is used as a fuel for domestic heating. Concentrations (and contributions to PM₁₀) were higher in background and residential areas, while traffic impacted sites experienced an additional PM₁₀ load from traffic emissions. The contribution of wood smoke to organic carbon and PM₁₀ levels was calculated using a constant ratio of levoglucosan and OC and PM₁₀ respectively which was derived for fire woods typical for Alpine European regions. For OC, levoglucosan concentrations are multiplied by 6.1 and for PM the factor is 10.7 (Schmidl et al. 2008a). The estimated contribution of wood smoke-OC to total OC in PM₁₀ ranged from one third to more than half in the cold season with higher contributions of up to 70% in winter (December, January and February) in the smaller cities and the rural background. Wood smoke can therefore be considered the predominant source of organic material at rural and small urban sites in central Europe. Consistently, wood smoke was an important contributor to PM₁₀ during the cold season, with contributions of around 10% in the larger Vienna region and around 20% at rural sites in the densely forested regions of Salzburg and Styria during the winter months. In those regions the highest relative wood smoke contents was found in residential regions during fall (September till November), when wood stoves are used as auxiliary heating systems in the transition from the warm to cold season. In alpine valleys wood smoke can account even for more than 50% of PM₁₀ (Bauer et al. 2010). Using the relationships between the different anhydrous sugars described in the method section, the combustion of softwood was found to be dominant for wood smoke at all sites.

Data on wood smoke emissions by different fuels as well as by different combustion systems and combustion conditions are important for emission inventories and source apportionment studies. Such studies were carried out by the Vienna University of Technology team (Luisser et al. 2008, Schmidl et al. 2008c, Gonçalves et al. 2010). Emission factors for particulate emissions from different wood types and combustion conditions were determined and the biomass smoke was chemically characterized. More than 50 chemical constituents were analyzed including carbonaceous species, soluble ions, anhydrous sugars, HULIS, n-alkanes, carboxylic acids, PAHs and trace metals.

5. References

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