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5.1 Sources of different carbon fractions of carbonaceous aerosols

Carbonaceous aerosols, composed of elemental carbon (EC) and organic carbon (OC), are major contributors to PM$_{2.5}$ mass concentrations (e.g., Liu et al., 2017; Cao et al., 2007). Understanding the sources and atmospheric formation processes of its major contributors is thus important for mitigation of PM$_{2.5}$ pollution.

EC is almost exclusively produced from incomplete combustion of biomass and fossil fuel. Thus, changes in isotopic composition of EC directly reflect changes in these emission sources. Figure 5.1 shows the relative contribution of non-fossil (i.e., biomass burning) and fossil sources (i.e., coal combustion, vehicle emissions) to EC by radiocarbon ($^{14}$C) measurement in 6 Chinese cities including 3 cities in northern China (Xi'an, Beijing, Taiyuan), where coal is widely used for heating in winter, and 3 in southern China (Shanghai, Chongqing and Guangzhou). The most important contributor to EC is fossil fuel combustion regardless of variabilities in different cities and seasonality, contributing from 60% in Chongqing to 90% in Taiyuan. This suggests considerable contributions of biomass burning to EC in Chongqing and that fossil sources dominate over biomass burning in Taiyuan. Taiyuan, known for its coal production, is ranked as the most air-polluted city in China. It is a typical Chinese city in terms of heavy reliance on coal as a primary source of heat and power. On the other hand, Chongqing is located in Sichuan Basin, which is the agricultural heartlands of the southwestern China. Chongqing is influenced by anthropogenic sources (e.g., biomass burning, vehicle emissions, industrial etc.) and surrounded by mountains which is unfavourable for dispersion of air pollution. For the 1-year samples collected in Xi’an during 2008/2009, fossil sources of EC are distinguished into coal combustion (brown bar) and vehicle emissions (or liquid fossil fuel combustion) by complementing radiocarbon with stable isotope $^{13}$C in a Bayesian Markov Chain Monte Carlo (MCMC) approach. The relative contributions from coal combustion and vehicle emissions are estimated as 45% (median; 29%–58%, interquartile range) and 31% (18%–46%) in winter, respectively, whereas in other seasons more than one half of EC is from vehicle emissions and coal burning is much less important. However, increased contributions from biomass burning (1.2µg m$^{-3}$) were important for the EC increment in winter (2µg m$^{-3}$) as well, contributing 60% of the EC increment in winter. Comparisons with other winter-time studies at the same sampling site suggest that the sources of fossil EC have changed from 2008/2009 (this study) to 2013/2014 (Wang et al., 2016; Zhang et al., 2015), with decreasing
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contributions from coal burning and increasing contributions from motor vehicles. This is consistent with recent changes in Xi’an: changes in energy consumption, and the expansion of the civil vehicular fleet resulting from urbanization and economic improvement.

Unlike EC that is exclusively emitted as primary aerosols, OC is either directly emitted from non-fossil (e.g., biomass burning, biogenic emissions, and cooking) and fossil sources as primary OC (POC), or formed in the atmosphere via gas-to-particle conversion as secondary OC (SOC). Compared with EC, OC has much higher contribution from non-fossil sources for all the cities and seasons. The main reason for this is that biomass burning emits more OC relative to EC than the fossil sources. So even if biomass burning contributes a small fraction to EC, it will have a much higher contribution to primary OC. Additionally other sources, such as primary and secondary biogenic emissions as well as cooking will contribute to OC, but not to EC. Secondary formation can contribute both fossil and non-fossil carbon to OC and it is not a priori clear which contribution will be larger. Of the fossil sources coal combustion has a higher primary OC to EC ratio than vehicle emissions. Therefore in a city where biomass burning and coal combustion are the dominant pollution sources, the fraction modern of primary OC and EC will be more similar than in a city where the main sources are biomass burning and traffic. If secondary formation is comparable for fossil or non-fossil sources this will hold for total OC as well. This can be illustrated by the formulas of the fraction non-fossil in EC and OC ($f_{bb}(EC)$ and $f_{nf}(OC)$, respectively; Eq. 5.1-5.3). Comparing Eq. (5.1) with Eq. (5.3), the differences between $f_{bb}(EC)$ and $f_{nf}(OC)$ depend on the differences between $r_{fossil}^{bb} \times \frac{1 + y_{fossil}^{bb}}{1 + y_{nf}^{nf}}$ and 1.

$$f_{bb}(EC) = \frac{EC_{bb}}{EC_{bb} + EC_{fossil}} = \frac{1}{1 + \frac{EC_{fossil}}{EC_{bb}}} \quad (5.1)$$

$$f_{nf}(OC) = \frac{OC_{nf}}{OC_{nf} + OC_{fossil}} = \frac{POC_{bb} + OC_{o,nf}}{POC_{bb} + OC_{o,nf} + POC_{fossil} + SOC_{fossil}} \quad (5.2)$$

$$f_{nf}(OC) = \frac{1}{1 + \frac{(1 + y_{fossil})}{(1 + y_{nf})} \times \frac{POC_{fossil}}{POC_{bb}}} = \frac{1}{1 + \frac{EC_{fossil}}{EC_{bb}} \times r_{fossil}^{bb} \times \frac{1 + y_{fossil}^{bb}}{1 + y_{nf}^{nf}}} \quad (5.3)$$

where POC_{bb} is the primary OC (POC) from biomass burning; POC_{fossil} is the
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POC from fossil fuel combustion including both coal combustion and vehicle emissions; OC_{nf} is the non-fossil OC (OC_{nf}) excluding POC_{bb} (e.g., OC_{o,nf} includes secondary OC from biomass burning, primary and secondary OC from biogenic emissions and cooking); SOC_{fossil} is the secondary OC from fossil fuel combustion; \( y_{\text{fossil}} \) is equal to SOC_{fossil} over POC_{fossil}; \( y_{\text{nf}} \) is equal to OC_{o,nf} over POC_{bb}; \( r_{\text{fossil}} \) is the POC to EC ratio for fossil sources; \( r_{\text{bb}} \) is the POC to EC ratio for biomass burning.

The smallest differences between the non-fossil fraction of OC and EC were found in Beijing and biggest differences in Chongqing in winter. For Beijing this suggests either strong contribution from coal combustion or that secondary formation strongly favors fossil OC, or both. For Chongqing, low contributions from coal combustion and/or non-fossil dominated SOC can explain the observations. Earlier studies on OC in Beijing found that fossil OC is dominated by coal combustion and is mostly secondary in nature, although the detailed atmospheric formation mechanisms responsible for the enhancement in fossil SOC formation remain unclear (Huang et al., 2014; Elser et al., 2016).

For the year 2008/2009, higher contribution of non-fossil sources (e.g., biomass burning, biogenic emissions, and cooking etc) to OC_{nf} (OC) was found in winter (62 \%) than in summer (48 \%) in Xi’an. A different seasonal variation pattern for \( f_{\text{nf}} \) (OC) was reported in Beijing, where the non-fossil contribution to OC was lower in winter than in summer (Yan et al., 2017; Zhang et al., 2017). The variability in the contribution of non-fossil and fossil sources to EC and OC as well as their different seasonal variations imply that different pollution patterns exist in different Chinese cities. Earlier source apportionment results found that during wintertime biomass burning is a major source of OC in Xi’an and the coal combustion dominate the OC mass in Beijing (Elser et al., 2016). Efforts to mitigate air pollution in China should be put into regulating the activities of biomass burning and coal combustion, and control strategies to improve air quality in local areas need to consider different pollution sources in different cities.

Recent studies have revealed that SOC is an important or sometimes the dominant contributor to high concentrations of PM_{2.5} in China (e.g., Huang et al., 2014a). But direct \(^{14}\text{C}\) measurements on SOC are not possible, as isolating the SOC from total OC is technically and conceptually difficult. In this thesis sources and formation mechanisms of SOC were characterized by the following approaches.
1) Estimating SOC concentrations and sources based on the $^{14}$C-apportioned OC and EC. This is applied to the PM$_{2.5}$ samples collected in 6 typical Chinese cities (i.e., Xi’an, Beijing, Taiyuan,
Shanghai, Chongqing, and Guangzhou) in winter 2014/2015. The estimated relative contribution of fossil sources to SOC ($f_{\text{fossil}}(\text{SOC})$) ranged from 10% in Chongqing to 70% in Beijing, averaging 43% (Fig. 5.2). $f_{\text{fossil}}(\text{SOC})$ is in general smaller than $f_{\text{fossil}}(\text{OC})$, expect in Beijing where $f_{\text{fossil}}(\text{SOC})$ is bigger than $f_{\text{fossil}}(\text{OC})$. In addition, the fossil OC in Beijing is predominately of secondary origin, with secondary formation accounting for 67% of fossil OC (i.e., $(\text{SOC}/\text{OC})_{\text{fossil}} = 67\%$), compared to 38% in other cities. This is also found in an earlier study that during winter 2013, where $(\text{SOC}/\text{OC})_{\text{fossil}}$ in Beijing was much higher than in Xi’an, Shanghai and Guangzhou (Zhang et al., 2015).

The highest absolute $\text{SOC}_{\text{fossil}}$ concentrations are also found in Beijing (25 μg m$^{-3}$ for haze days in Beijing, 5 times higher than averaged $\text{SOC}_{\text{fossil}}$ in other cities (i.e., Xi’an, Taiyuan, Shanghai and Guangzhou). Concentrations of SOC from non-fossil sources (SOC$_{nf}$) ranged from 2.0 μg m$^{-3}$ found in Guangzhou to 18.1 μg m$^{-3}$ in Chongqing, with an average of 8.5 ± 5 μg m$^{-3}$. SOC$_{nf}$ is a significant fraction of the total non-fossil OC (OC$_{nf}$), ranging from 38% to 73%. The contribution of secondary sources to OC is on average 16 ± 8% higher (absolute) for non-fossil OC than for fossil OC in most places we studied (Xi’an, Taiyuan, Shanghai, Chongqing and Guangzhou). On the contrary, Beijing has lower $(\text{SOC}/\text{OC})_{nf}$ than $(\text{SOC}/\text{OC})_{fossil}$ with an average absolute difference of 13%. Beijing seems to be a unique place with very large importance of secondary OC from fossil sources. This can be seen both from the high fossil fraction of SOC and that high contribution of secondary sources to fossil OC, which exceeds even that of non-fossil OC. The reason for why Beijing is different from other cities also influenced strongly by coal burning is not clear so far, and deserves further study.

2) **Comparing $\delta^{13}\text{C}_{\text{OC}}$ with $\delta^{13}\text{C}_{\text{EC}}$.** The $\delta^{13}\text{C}_{\text{EC}}$ preserves the signature of emission sources, but OC is chemically reactive and $\delta^{13}\text{C}$ signatures of OC are not only determined by the source signatures but also influenced by atmospheric processing. This is applied to the one-year PM$_{2.5}$ samples in Xi’an, China in 2008/2009. $\delta^{13}\text{C}_{\text{OC}}$ exhibited similar values with $\delta^{13}\text{C}_{\text{EC}}$, and showed strong correlations ($r^2 = 0.90$) in summer and autumn, indicating similar source mixtures with EC. In spring, $\delta^{13}\text{C}_{\text{OC}}$ is depleted (1.1–2.4‰) compared to $\delta^{13}\text{C}_{\text{EC}}$, indicating the importance of secondary formation of OC (e.g., from volatile organic compound precursors). SOA is produced by the oxidation of gaseous precursors,
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which proceeds faster for molecules depleted in heavy isotopes, leading to depleted reaction products relative to the precursors. These reaction products are involved in the formation of SOA and a depleted OC signature can therefore be an indication of secondary formation from incompletely oxidized precursor gases. According to our data SOA formation seems to be especially important in springtime.

3) **Comparison of concentrations and sources of POC with those of total OC.** This approach makes it possible to provide some insights into the importance of secondary formation and other chemical processes, such as photochemical loss mechanisms. Because the observed total OC also includes secondary OC from fossil fuel combustion (i.e., coal combustion and vehicle emissions) and non-fossil sources (e.g., biomass burning, biogenic emissions), in addition to POC. This is also applied to the one-year PM$_{2.5}$ samples in Xi’an China in 2008/2009. Primary OC mass is estimated by multiplying EC concentrations attributed to each source by the corresponding primary POC/EC emission ratios. The estimated primary OC is similar to the observed total OC concentrations except for samples with observed OC concentrations >25 μg m$^{-3}$. POC concentrations also show similar variability and seasonality as the total OC concentrations ($r^2 = 0.71$), However, the non-fossil fraction in estimated primary OC is significantly lower than the observed $f_{nf}(OC)$ and the non-fossil fractions of total OC and primary OC are not well correlated ($r^2 = 0.31$). Those differences can be explained by the contribution of other non-fossil primary OC (excluding biomass burning), or secondary non-fossil OC, which are not included in the estimation. But we cannot reconcile the differences between observed and estimated non-fossil OC fraction without overestimating the absolute OC concentrations, especially in summer. Therefore, we hypothesize that loss of primary OC due to active photochemistry cannot be neglected, especially not in summer.
Newly formed secondary OC (SOC) and recently emitted primary OC are usually more volatile than OC that has undergone extensive photochemical processing in the atmosphere (aged OC). Therefore, understanding the sources of more and less volatile fraction of OC is of great importance for better understanding the complex sources and atmospheric processes of organic aerosols. We thus investigate for the first time the sources of more volatile OC (mvOC) by radiocarbon measurement. Aerosol samples collected in 6 Chinese cities during wintertime were desorbed in He at 200 °C to isolate mvOC. Even...
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though the desorption temperature is not a direct measure of the particle volatility, OC desorbed from the filter at lower temperature tends to be more volatile, and the fraction of mvOC in the total OC (mvOC/OC) provides an indication of OC volatility. The 6 Chinese cities have an average mvOC/OC of 10.5 ± 3.3 %, smaller than ambient samples from other places and much smaller than fresh source samples (Fig. 5.3). This suggests active atmospheric processing of the emitted OC during wintertime in urban China. The fossil contribution to mvOC is consistently larger than that to SOC and OC (Fig. 5.2a), regardless of their spatial variability in urban China, suggesting that mvOC is influenced more strongly by fossil sources than SOC and OC. The fossil fraction of mvOC is related to primary OC emissions from fossil sources, indicated by strong correlation between mvOC and primary OC from fossil sources (mvOC_{fossil} vs. POC_{fossil}; $r^2 = 0.69$) and no correlation between mvOC and secondary OC from fossil sources (mvOC_{fossil} vs. SOC_{fossil}; $r^2 = 0.01$). Correlation between mvOC and secondary OC from non-fossil sources (mvOC_{nf} vs. SOC_{nf}; $r^2 = 0.88$) is somewhat stronger than mvOC and primary OC from non-fossil sources (mvOC_{nf} vs. POC_{bb}; $r^2 = 0.71$). This indicates that secondary formation of OC from non-fossil sources (e.g., biomass burning, biogenic emissions, cooking) explains slightly more of the variation in mvOC_{nf} than primary biomass burning.

Figure 5.3. The mvOC fraction in total OC (mvOC/OC) of ambient samples collected in 6 typical Chinese cities (Xi’an, Beijing, Taiyuan, Shanghai, Chongqing and Guangzhou) during wintertime (this study), ambient mvOC/OC in other locations (Prague; Vodička et al., 2015), mvOC/OC of fresh and aged OC in chamber study from biomass burning (Keller and Burtscher, 2017). For fresh emissions of vehicle (Zhu et al., 2014, 2010) and coal combustion (Tian et al., 2017), the contribution of OC desorbed up to 140°C, and 280°C to total OC are shown as blue square and red square, respectively. The mvOC/OC thus would fall in between the blue and red square.
Concentrations of SOC are consistently higher than that of m vOC. SOC$_{fossil}$ is on average 4 times higher than m vOCS$_{fossil}$ and SOC$_{nf}$ 7 times higher than m vOCS$_{nf}$ (Fig. 5.2). This shows that most SOC from both fossil and non-fossil sources has desorption temperature above 200 °C, likely due to atmospheric aging of newly formed SOC leading to aged SOC.

Higher fossil contribution to m vOC than to OC in the 6 Chinese cities in this study is unambiguously concluded from $^{14}$C source apportionment. It is consistent with conclusions from $\delta^{13}$C of OC desorbed at different temperatures for ambient samples in other locations (i.e., urban, coastal and forest site in Lithuania; Masalaite et al., 2017, 2018). However, the conclusions from $\delta^{13}$C of Masalaite et al. (2017, 2018) remained qualitative, because $\delta^{13}$C of OC can be changed significantly by isotopic fractionation during atmospheric processing. The $^{14}$C source apportionment are thus more straightforward compared to $\delta^{13}$C.

The source endmembers for $\delta^{13}$C are less well-constrained than for $F^{14}$C, as $\delta^{13}$C varies with fuel types and combustion conditions. For example, $\delta^{13}$C values for liquid fossil fuel combustion has some overlap with $\delta^{13}$C values for both coal and C3 plant combustion. $^{13}$C source apportionment of fossil EC (EC$_{fossil}$) into EC from coal combustion (EC$_{coal}$) and vehicle exhaust (EC$_{vehicle}$) is therefore very uncertain. $\delta^{13}$C source signatures for EC are fully complied and established by a thorough literature search, but there are not many studies on $\delta^{13}$C of EC from different combustion sources and how it changes with combustion conditions. We did not further distinguish the contribution of coal combustion and vehicle exhaust in fossil OC, because OC is chemically reactive, and $\delta^{13}$C of OC can be affected by atmospheric processing (Kirillova et al., 2013). The stable carbon isotope composition ($\delta^{3}$C) of OC changes according to the kinetic isotope effects (KIEs) associated with various chemical reactions involved in aging of OC and the secondary formation from volatile organic precursors. For further studies, $\delta^{13}$C of EC from localized emission sources are in urgent need to better separate EC$_{coal}$ from EC$_{vehicle}$. Further, understanding of the KIE that a reactant and its products undergo is necessary for improving our knowledge of OC sources and formation mechanisms.

Estimation of POC and SOC, based on apportioned OC and EC and the corresponding POC/EC ratios for different sources, is limited by the uncertainties regarding the knowledge of POC/EC ratios for different sources, which are variable with burning condition and even measurement methods. For example, if all source ratios of POC/EC are overestimated, then POC calculated by multiplying EC with POC/EC ratio will also be overestimated. SOC,
calculated by subtracting POC from total OC, will be thus significantly underestimated. Or, if EC_{coal} is overestimated compared to EC_{vehicle}, then POC from coal (POC_{coal}) will increase and POC from vehicle exhaust (POC_{vehicle}) will to a less extent decrease, due to higher POC/EC ratio for coal combustion than that for vehicle emissions. This will lead to SOC being significantly underestimated as well. Because of the importance of POC/EC source ratios in estimating POC and SOC, further studies need to be carried out to understand the variation of POC/EC ratio with combustion conditions, sampling artefacts, measurement technique of OC and EC, and meteorological conditions etc., and finally get the representative POC/EC source ratios. In addition, residential coal combustion is one of the major contributors to severe air pollution in some regions (e.g., China, India, Ireland, Eastern Europe, but knowledge on its emission characteristics of primary OC and EC are still limited compared to biomass burning and vehicle exhausts. More studies on emissions of residential coal combustion are welcomed in the literature.

The isotope source estimation for SOC cannot provide direct information on the formation mechanism of SOC. It would be more powerful by integration of isotopic and chemistry studies (e.g., Zhang et al., 2017; Vlachou et al., 2018). For example, coupling $^{14}$C analysis with the positive matrix factorization (PMF) analysis of aerosol mass spectrometer data can provide more detailed information of the OC sources including identification of SOC based on OC precursors than either method alone. Vlachou et al. (2018) applied this method to the organic aerosol in the Alpine valley of Magadino for the year 2013-2014, and found that SOC was formed mainly by biogenic sources during summer and anthropogenic sources during winter.

Moreover, OC can be separated into water-insoluble OC (WIOC) and water-soluble OC (WSOC) by water-extraction. Fossil sources tend to mainly produce WIOC (e.g., Huang et al., 2014b; Yan et al., 2017; Dai et al., 2015) in primary emissions. Therefore, fossil WIOC can be used as a proxy of primary fossil OC. WSOC can be directly emitted as primary aerosols mainly from biomass burning or produced as SOC. There are evidences that SOC produced through the oxidation of volatile organic compounds followed by gas-to-particle conversion contains more polar compounds and thus may be an important source of WSOC (Sannigrahi et al., 2006; Kondo et al., 2007; Weber et al., 2007; Miyazaki et al., 2006). Fossil WSOC therefore thought to be a good proxy of fossil SOC. $^{14}$C analysis of WIOC and WSOC can therefore provide new insights into sources and formation processes of primary and secondary OC, respectively (e.g., Dusek et al., 2017; Liu et al., 2016).
5.2 Light absorption of primary and aged coal combustion emissions

Recent studies reveal that besides biomass burning, coal combustion is also an important source of light-absorbing organic aerosols (referred to as brown carbon, BrC). However, our understanding of BrC from coal combustion and how the atmospheric processing modifies the light absorption of BrC is still very poor. This limits climate models in their capacity to take into consideration of BrC absorption in radiative forcing estimates.

In this work, we present, to our best knowledge, the first light absorption measurement of BrC extracts from primary and aged residential coal combustion emissions based on controlled smog chamber experiments. We conduct 12 individual burns in a traditional Chinese stove commonly used in the residential sector, using 3 bituminous coals (B1, B2 and B3) and 2 anthracite coals (A1 and A2) collected from major coal producing areas in China. The bulk mass absorption coefficient (bulk MAC) of primary and aged coal combustion OA in methanol extracts were determined and used to represent the total light absorption ability of coal burning BrC, because OA from coal combustion is largely water-insoluble. The bulk MAC of methanol extracts varies by coal type and burns, and is dependent on the combustion conditions indicated by \( f_{OA} \) (i.e., \( \frac{OA}{(OA+BC)} \)). We compared the methanol-extracted bulk MAC of coal burning with those of biomass burning and gasoline vehicle emissions and suggests that in addition to biomass burning, residential coal combustion and gasoline vehicle emissions could also play an important role in light absorption of BrC and should be considered in further modeling of OA radiative forcing. We found decreased bulk MAC with aging, suggesting that the SOA condensing on the primary particles is only weakly light absorbing.

Compared to primary OA, bulk MAC of aged OA (POA + SOA) is much less variable, which may arise from the production of secondary BrC or the transformation of primary BrC with aging. We conclude that the SOA has to be light absorbing to explain the decreased bulk MAC of aged OA compared to primary OA. The bulk MAC of SOA was estimated based on the bulk MAC of aged OA and primary OA, and we found that bulk MAC of SOA is related to the SOA formation and is affected by combustion conditions.

Although studies of the bulk light absorption of aerosol extracts have been used to investigate the sources, extent, and chemistry of fine particulate BrC, it is not straightforward to use this method to assess optical properties of airborne BrC particles. To convert the solution bulk MAC to light absorption by an aerosol, knowledge of both particle morphology and how the chromophores are
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distributed at different particle size is needed. For example, Liu et al. (2013) found that the light absorption of ambient BC is higher than the light absorption of the bulk solution by a factor of 2 for water extracts and by a factor of 1.8 for methanol extracts. However, people often chose bulk MAC to communicate, because the bulk MAC can be used to derive the imaginary refractive index (i.e., $k$):

$$
 k = \frac{\rho \times \lambda \times \text{bulk MAC}(\lambda)}{4\pi}
$$

(5.4)

where $\rho$ is the density of the dissolved compounds. A $\rho$ must be chosen to obtain an imaginary refractive index ($k$) from bulk MAC. The $\rho$ is somewhat uncertain, but it is only an intermediate value; it is required again in Mie or other models that calculate particulate absorption per mass. If the same $\rho$ is used in both calculations, it has little effect on particulate absorption.

Further studies of BC should target the identification of the specific chromophores (i.e., components of molecules that are responsible for light absorption by BC) responsible for the color change during aging processes. To date, the link between BC’s molecular composition and light absorption properties remains poorly known, because the identification of BC chromophores is still a challenge. This is due to the low concentrations of light-absorbing molecules in complex organic mixtures. It is thus necessary to develop a highly sensitive molecular characterization approaches capable of detecting small changes in the chemical composition of BC.

In the atmosphere, the BC is contributed by different sources (e.g., biomass burning, coal combustion, vehicle emissions, biogenic emissions etc.). Additional work is needed to establish the link of different emission sources to the total OA absorbance, which is of great importance to improve the knowledge on the climate forcing of aerosol. The class of humic-like substance (HULIS) in aerosol has been found to strongly link to atmospheric BC (Lukács et al., 2007), and can be chemically isolated from filter samplers. Radiocarbon measurements of HULIS will be powerful, for which the relative contributions of fossil fuel combustion versus biomass burning/biogenic sources to HULIS can be clearly differentiated.
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