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Inferring absorbing organic carbon content from AERONET data

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Abstract. Black carbon, light-absorbing organic carbon (often called "brown carbon") and mineral dust are the major light-absorbing aerosols. Currently the sources and formation of brown carbon aerosol in particular are not well understood. In this study we estimated globally the amount of light-absorbing organic carbon and black carbon from AERONET measurements. We find that the columnar absorbing organic carbon (brown carbon) levels in biomass burning regions of South-America and Africa are relatively high (about 15-20 mg/m^2 during biomass burning season), while the concentrations are significantly lower in urban areas in US and Europe. However, we estimated significant absorbing organic carbon amounts from the data of megacities of newly industrialized countries, particularly in India and China, showing also clear seasonality with peak values up to 30-35 mg/m^2 during the coldest season, likely caused by the coal and biofuel burning used for heating. We also compared our retrievals with the modeled organic carbon by global Oslo CTM for several sites. Model values are higher in biomass burning regions than AERONET-based retrievals, while opposite is true in urban areas in India and China.

1 Introduction

Aerosols affect both directly and indirectly the Earth's climate. Currently, aerosol forcing is the largest uncertainty in assessing the anthropogenic climate change (IPCC, 2007). Recent research (e.g., Myhre, 2009) has emphasized that in addition to aerosol optical depth (AOD), assessments of aerosol effects on climate require globally distributed information on aerosol characteristics such as size, composition and optical properties, particularly absorption. Moreover, it

is of importance to get better understanding of the different absorbing components.

Black carbon (BC) is the main absorbing component present in atmospheric aerosols. In addition, soil dust absorbs light in the UV and visible, and some organic materials absorb in the UV. The enhanced UV absorption feature was observed for instance by Martins et al. (2009), who analyzed data from Sao Paulo. Kirchstetter et al. (2004) showed strong spectral dependence on the light absorption by organic aerosols in the UV. Similar absorption characteristics, attributed to organics, have been found in several other reported measurements [e.g. Yang et al. (2009); Alexander et al. (2008); Lewis et al. (2008)], also in Russell et al. (2010) and references therein. The sources and formation of light-absorbing organic carbon compounds (often called as "brown carbon") and their temporal variability, however, are currently not well understood (Andreae and Gelencsér, 2006; Bergstrom et al., 2007). Moreover, their optical properties change during aging (Laskin et al., 2010).

Recently, Schuster et al. (2005, 2009) have introduced a methodology to derive information about both aerosol black carbon content and water uptake from AEROSOL ROBOTIC NETWORK (AERONET)-retrieved complex refractive index. Koven and Fung (2006) separated the absorption by black carbon from absorption by dust, applying the wavelength-dependent absorption in AERONET data. Koch et al. (2009) compared modeled BC concentrations against available measurements (including an updated version of Schuster et al. (2005) data set) and found underestimation of BC by global aerosol models and relatively large diversity between different models. Model behavior (including the emissions) against OC estimates has not been compared to measurements due to the lack of a global reliable OC surface or columnar data set. Sato et al. (2003) focused on OC in addition to BC, but they still constrained the modeling by aerosol absorption optical depth from AERONET, not attempting to retrieve directly OC concentrations from AERONET data.

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Russell et al. (2010) assessed the usefulness of AERONET-retrieved single scattering albedo (SSA), aerosol absorption optical depth (AAOD), and absorption Ångström exponent (AAE) as indicators of aerosol composition, including black carbon, organic matter, and mineral dust. They did not, however, estimate the composition from AERONET measurements.

In our study reported herein, the approach of Schuster et al. (2005) was extended and refined to assess whether globally relevant information about the relative role of organic carbon (OC) absorption can be inferred from AERONET measurements. We retrieved the columnar mass concentration of absorbing organic carbon in mg/m^2 and OC/BC ratio from all available AERONET Level-2 data. We find that the levels in biomass burning regions of South-America and Africa are relatively high, while the concentrations are significantly lower in urban areas in US and Europe. However, significant absorbing OC amounts were estimated from the data of megacities of newly industrialized countries, particularly in India and China during coldest season.

2 Data and methods

2.1 AERONET data

AERONET (AErosol RObotic NETwork) is a globally distributed network of automatic sun and sky scanning radiometers that measure at several wavelengths, typically centered at 0.34, 0.38, 0.44, 0.50, 0.67, 0.87, 0.94, and 1.02 μm . Each band has a full width of approximately 0.010 μm at half maximum (FWHM). All of these spectral bands are utilized in the direct Sun measurements, while four of them are used for the sky radiance, 0.44, 0.67, 0.87 and 1.02 μm . Spectral aerosol optical depth (AOD) is obtained from direct sun measurements, and inversion products of other aerosol optical properties, such as single scattering albedo (SSA), refractive indices and the column integrated aerosol size distributions above the measurement site are provided at the sky radiance wavelengths (Holben et al., 1998).

In our study, we used level 2.0 data (quality assured) of refractive indices at four retrieval wavelengths 0.44, 0.67, 0.87 and 1.02 μm to match model derived refractive indices, in order to retrieve black carbon and organic carbon concentrations.

2.2 Retrieval of absorbing OC

Schuster et al. (2005) demonstrated an approach to retrieve black carbon concentration and specific absorption from AERONET retrievals of imaginary refractive indices. Dey et al. (2006) applied this same approach, but included additionally OC or dust as absorbing component. However, they did not focus on retrieved OC concentrations, which is the main focus of our study. We utilized the AERONET measurements of refractive index, and refined the approach of

Schuster et al. (2005) to focus on spectral absorption and possible influence of OC, to assess whether this kind of information can be retrieved to have climatologically and globally relevant information. Since the main details of the methodology are comprehensively described elsewhere, particularly in Schuster et al. (2005), only main points and assumptions used in our study are summarized here.

We applied Maxwell-Garnett (MG) mixing rule for a mixture of BC, OC and $(NH_4)_2(SO_4)$ embedded in water host. The assumed refractive index values of these components are given in the Table 1. We used the values for BC refractive index and density suggested by Bond and Bergstrom (2006). We considered a spectrally independent imaginary part, $k(\lambda)$ except for OC, for which we fixed $k(\lambda)$ at 440 nm and 670 nm with the values used by Kirchstetter et al. (2004). There are lot of different OC species with varying absorption properties. However, many of the organic species do not absorb significantly and it has to be stressed that in our approach “organic carbon” should be taken as equivalent for concentration of components causing enhanced absorption at the shortest wavelengths that cannot be explained by BC.

From MG, $k(\lambda)$ were computed for different combinations of volume fractions of BC (f_{BC}) and OC (f_{OC}) by minimizing the following equation:

$$\chi^2 = \sum_{i=1}^4 \frac{(m_i^{rtv} - m_i^{cal})^2}{(m_i^{rtv})^2}, \quad (1)$$

where m_i^{rtv} is the AERONET-retrieved imaginary index and m_i^{cal} is the value based on MG mixing rule, i is the summation index over four wavelengths from AERONET.

The absorbing OC concentration is calculated then

$$[OC] = f_{OC} * \rho_{OC} * \int \frac{dV}{d \ln r} d \ln r, \quad (2)$$

where f_{OC} is volume fraction of absorbing OC that gave the best fit in the equation 1, $\frac{dV}{d \ln r}$ is the particle volume distribution (in $\mu m^3 \mu m^{-2}$), available from the AERONET inversion product. For a density of OC (ρ_{OC}), we used the same value of 1.2 $g \text{ cm}^{-3}$ that was suggested by Turpin and Lim (2001).

In order to exclude dust dominated cases, we processed only those retrievals that had the ratio of fine mode to total volume concentration larger than 0.5. Moreover, we included the spectral imaginary index with the following restriction, AERONET-based imaginary index at 440nm had to be larger than the mean of the imaginary indices at 440, 670 and 1020 nm.

Figure 1 shows an example of our absorbing OC retrievals for several sites as a function of AAE and fine mode to total volume concentration (FMF). AAE is calculated from 440 nm and 670 nm wavelength pair. It can be noted that the mean FMF in Kanpur and Beijing is smaller compared to the other sites. On average, an increase in AAE relates to stronger increase in the retrieved absorbing OC amount in

Kanpur and Beijing than in biomass burning dominated sites. This is caused by both f_{OC} and total volume concentration (in equation 2) being larger for any given AAE in these sites. Russell et al. (2010) and Shinozuka et al. (2009) showed similar relationships between AAE and total organic mass fraction for data over Central Mexico. However, in our case the relationship should be more evident, since we do not include total but absorbing OC.

3 Results

We included all the AERONET sites and calculated the monthly means of different retrieved parameters at 5x5 degree boxes. This means that in some regions several sites are included within one 5x5 degree box, particularly in Europe and US. In addition, it was required that each month includes at least 10 measurements. We also stress that since we used Level-2 data, our retrievals are essentially restricted to the cloud-free cases with AOD at 440nm larger than 0.4 and solar zenith angle larger than $> 50^\circ$. This will bias the concentrations towards high values in many locations and months.

Schuster et al. (2005) did not include OC as a possible absorbing component, and estimated that this causes a positive bias of less than 9% in their retrieval of black carbon, when organic carbon aerosols are present. Therefore, by including organic carbon in this retrieval not only allows estimates for absorbing OC, but it leads to more accurate BC concentrations. Koch et al. (2009) compared modeled BC concentrations against available measurements (including an updated version of Schuster et al. (2005) data set) and found underestimation of BC by global models and relatively large diversity between different global aerosol models. Part of this can be attributed of systematic bias caused by neglecting wavelength dependence in AERONET- retrieved absorption. However, this is not a major explanation, since the difference between modeled and measurement-based BC was much larger in many cases.

Figure 2 shows the retrieved monthly mean absorbing organic carbon column concentration (in mg/m^2) in September. During this month, that includes most data and sites, the values of absorbing OC concentration are very similar in US and Europe (around 2-6 mg/m^2), while in the biomass burning regions of South America and South Africa they are typically higher and around 10-15 mg/m^2 . There are also elevated concentrations retrieved in the Top End of the Northern Territory of Australia in September that is within the burning season [e.g. Luhar et al. (2008)]. Qin and Mitchell (2009) classified a large part of the AERONET data as smoke in this region during the burning season, but also with class they called "super-absorptive". This class was distinct with very low single scattering albedo (0.7 at 500 nm) and enhanced absorption at 440 nm. They argued that the most likely source for these "super-absorptive" particles is the burning

of Australian hummock grasses (*Spinifex*), with high levels of both black and organic carbon.

The data from Bonanza Creek in Alaska and also from Moscow and Zvenigorod shows relatively high values, particularly if compared to the BC levels (OC/BC will be shown in the next figure). The grid-box including Beijing and Xi-anHe shows increased concentrations in China, however this region exhibits the highest values during winter months as shown and discussed later.

Figure 3 shows the seasonal variation of monthly mean absorbing OC and OC/BC at several sites located in very different aerosol environment. The left hand side panel includes sites with dominant biomass burning emissions, while right hand side has mainly urban sites with varying emission sources. Highest monthly mean value of OC/BC is in Bonanza Creek. These OC and BC emissions are mostly from boreal forest fires, OC/BC ratios from boreal forest fires are higher than from extratropical vegetation fires [e.g. Andreae and Merlet (2001)]. The imaginary refractive index of the Alaska smoke observations, analyzed by Eck et al. (2009), showed low values implying small BC fraction, and a very large relative increase towards lower wavelengths, likely resulting from enhanced absorption by OC, which is evident also in our retrievals. From the biomass burning sites, Alta Floresta exhibits somewhat larger OC/BC ratio than Mongu. Alta Floresta is mainly surrounded by tropical forest and to less extent by savanna (Cerrado), while Mongu is mainly surrounded by savanna, evident for instance by the proportions of vegetation types presented in Ichoku et al. (2008). Cuiaba Miranda is in the transition zone to Cerrado with less primary forest around than in Alta Floresta, however it exhibits rather similar absorbing OC concentrations. Value for OC/BC based on emission estimates of OC and BC by Savanna type, given in Andreae and Merlet (2001), is about 7.1, while it is 8.0 for Tropical Forest. Given the uncertainties involved in these estimates, the difference is very small, however it is in the same direction than our retrievals of OC/BC for Mongu and Alta Floresta. Reid et al. (2005) also reviewed observations on biomass burning emissions and gave ranges for BC/OC (in their Table 2) for different vegetation types. OC/BC for Savanna type (e.g. Mongu) ranges from 4.3-16. The values for boreal forest (e.g. Bonanza Creek) range from 12.5-33. Therefore, against these estimates too, our retrievals are consistent with the in-situ measurements conducted in these environments.

Retrievals in Kanpur and Gandhi College give relatively high absorbing OC concentrations for October-December period, when very high biomass burning (BB) occurs in this season (Tripathi et al., 2005a; Ganguly et al., 2009). Surface measurements of OC (Ram et al., 2010) in Kanpur, taking a boundary layer thickness of about 1 km (Tripathi et al., 2005b) and assuming that most OC reside within boundary layer, give a columnar OC of 38 mg/m^2 for October-December which is somewhat higher than predicted by the present retrieval, moreover their data show highest OC in

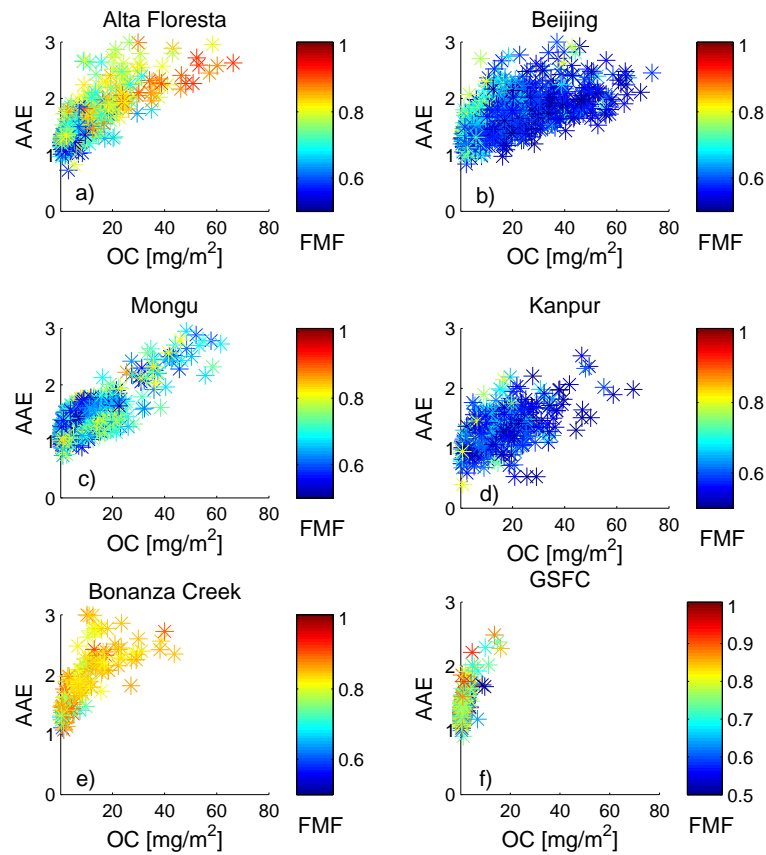


Fig. 1. Absorbing OC column concentrations [mg/m^2] as a function of AAE from 440/670 nm wavelength pair and the ratio of fine mode to total volume concentration (FMF). Name of the site is indicated at the top of each subplot.

December, while our retrieval gives highest absorbing OC amount for November.

The retrieved OC concentrations in some areas are affected strongly also by urban pollution (e.g. Beijing, Kanpur) are higher than those influenced mainly by biomass burning (e.g. Zambia, Brazil). This may seem first unexpected, however not much is currently known about brown carbon emissions from residential biofuels (Venkataraman et al., 2005) or from coal-burning aerosols, for instance, abundantly emitted particularly in these urban areas. Coal in China and other developing countries is widely used and furthermore, in many cases purposefully burnt inefficiently, to prolong the burning time (Yang et al., 2009), thus a further increase in brown carbon amount is a likely consequence. Cao et al. (2007) reports seasonal surface OC measurements in Beijing that are consistent with Lin et al. (2009), we also observe similarly strong seasonality with largest values in winter. Lin et al. (2009) explained their high winter-time OC concentrations by significant coal burning during the heating period,

particularly in rather close proximity to Beijing AERONET site. Although we tried to exclude the absorption by minerals, it has to be recognized that dust contamination is still possible in our retrievals to some extent. For example, Han et al. (2007) showed some rather high concentrations of mineral aerosols in the fine mode during the winter and spring in Beijing, and smaller amounts in the summer and fall.

Table 2 includes the comparison of monthly mean OC against modeling result from Oslo CTM-2 model (Myhre et al., 2009; Myhre, 2009). The model-based monthly mean OC data were taken from the cases when AOD at 440nm was larger than 0.4, to be consistent with this same requirement in the AERONET Level-2 data. In the OC retrievals we have used total volume concentration (equation 2), in other words equal fractions of organic carbon in the fine and coarse modes. Schuster et al. (2005) made this same assumption and moreover justified this being consistent with AERONET inversion where all particles have the same refractive index (Dubovik and King, 2000). However, in the Table 2 we also

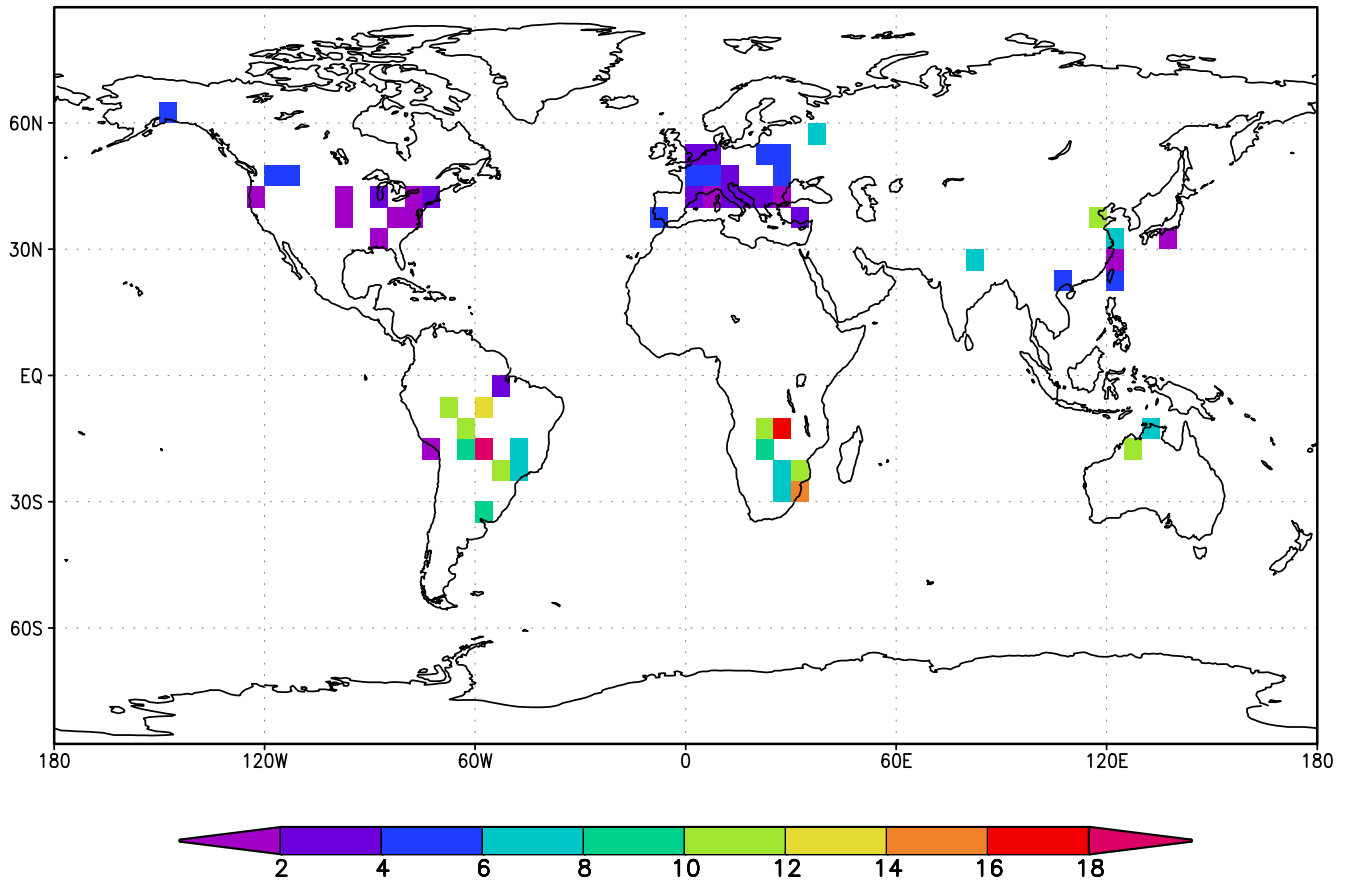


Fig. 2. Mean absorbing OC concentration (mg/m^2) inferred from AERONET-retrieved imaginary indices for September.

include the OC retrievals assuming volume concentration of fine mode only (the value in parenthesis).

The model peak months seem to take place earlier in many of the sites compared to AERONET-based values, which is more evident in Beijing and Kanpur than in BB sites. For instance, in Kanpur the model-based peak values occur in September-October, while highest AERONET-based absorbing OC concentrations are in October-November. The model gives higher OC concentrations in both biomass burning sites if compared to our AERONET-based retrievals. One has to keep in mind that model values take into account all the organic compounds, while our approach effectively gives "absorbing OC"; by definition components causing enhanced absorption at the shortest wavelengths. Measurement-based BC data, applicable to model comparison are sparse, which makes the effort by Koch et al. (2009) very valuable study, also to evaluate the black carbon emissions applied by the models. The column data for OC are even more sparse, so the main motivation for our study was to establish such a data set from the AERONET measurements. Although there

are significant uncertainties involved in our approach, mainly due to the fact that there exist numerous organic species with varying absorption characteristics (i.e. imaginary indices that are not well known), we want to emphasize that all the AERONET retrievals were consistently processed. Therefore, arguably the relative patterns, i.e. differences between regions and temporal variability in any given site, likely reveal more about the quality of the model emissions than absolute levels. Even if assuming fine mode concentration only in our retrieval, the model-based OC concentrations in Beijing and Kanpur are lower if compared to our retrievals. Underestimation in absorption optical depth was found in Sato et al. (2003) for these sites by both models that they include. They did not consider separately the influence by OC, while they increased both BC and OC to retrieve the best match between two global models and AERONET data.

There are not many measured refractive indices for OC species available at the wavelengths we are restricted to use now with the AERONET measurements. For instance, Kanakidou et al. (2005) (in their Figure 7) give optical con-

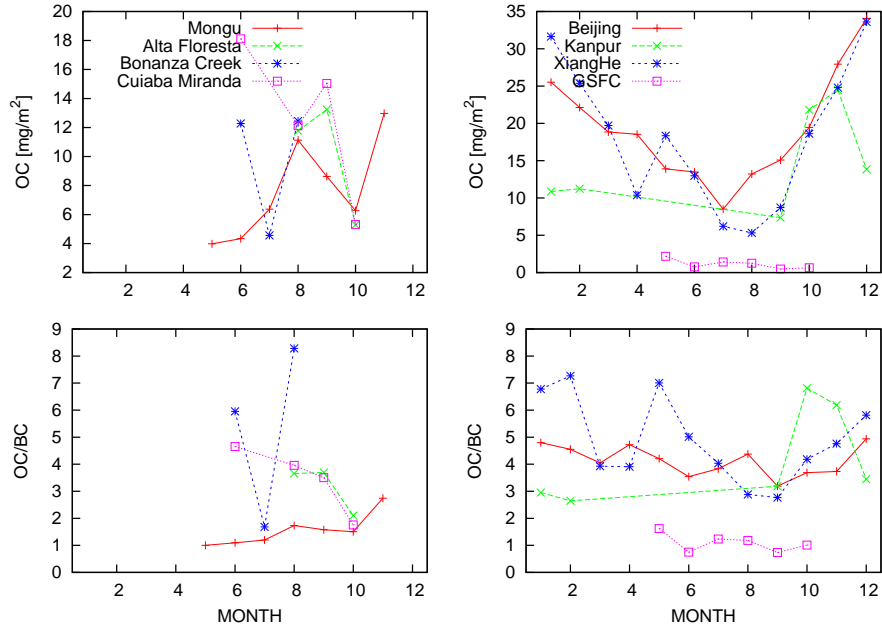


Fig. 3. Monthly mean absorbing OC [mg/m^2] for several biomass burning and urban sites.

stants of several organic components, most of which taken from Jacobson (1999), but all these imaginary indices are around 350 nm or lower. However, in order to carry out a sensitivity of our retrieval approach, we estimated absorbing OC concentrations using imaginary index values reported by Sun et al. (2007) and Chen and Bond (2010). Chen and Bond (2010) values are smaller than those of Kirchstetter et al. (2004) used in our study, therefore our retrieved absorbing OC concentrations in turn are lower. By using the refractive indices from Sun et al. (2007) and Chen and Bond (2010) we estimated the sensitivity of our approach to the assumed imaginary index values. We inferred the imaginary index from $\frac{\alpha}{\rho}$ values provided by Chen and Bond (2010) and Sun et al. (2007), assuming the same density we used in our retrieval (1.2 g cm^{-3}). Table 1 shows the imaginary index values at 440 nm and 670 nm from Sun et al. (2007) and Chen and Bond (2010) that we used in our sensitivity study. Figure 4 shows the monthly mean absorbing OC concentrations for Mongu with the two different imaginary index spectra from Chen and Bond (2010), for 210C and 360C production temperatures and two values from Sun et al. (2007), for water soluble organic carbon (WSOC) and water-insoluble organic carbon (WISOC). The relative changes in absorbing OC, due to these same different refractive index values, are similar in any other site. It is clear that the retrieved absorbing OC concentrations are sensitive to the assumed imaginary index. If lower values of imaginary index for 440nm and 670nm are used in our retrieval instead of Kirchstetter et al. (2004), significantly higher absorbing OC concentrations are retrieved for these biomass burning sites. In an ideal situations, there

were regionally varying and ecotype dependent imaginary index values in our global retrieval. Unfortunately, currently there are not sufficient data for this purpose. However, with the sensitivity study we wanted to highlight the importance of the imaginary index data in this kind of approach.

4 Conclusions

We have used global AERONET aerosol spectral refractive indices following an improved methodology of Schuster et al. (2005) to obtain columnar absorbing organic carbon (OC) and black carbon contents. One of the main assumptions in this approach is that higher imaginary index values at two shortest wavelengths (440nm and 670nm) are attributed to the absorption by organics (brown carbon), when the dust influence is attempted to exclude. In terms of AAE, this means that AAE higher than one (which is the estimated value for BC absorption) is due to the light-absorbing organic carbon. The actual retrieved absorbing organic carbon concentration then depends substantially on the assumed imaginary index for 440nm and 670nm, as was shown in the Figure 4. As an additional uncertainty, it has to be also noted that in some conditions the deviation from AAE=1 could be partly due to the lensing effect of scattering shell in BC core, as demonstrated by Lack and Cappa (2010). However, given the current uncertainties in our knowledge of formation and absorption properties of organic carbon and lack of global data of absorbing organic carbon, new approach to estimate both regional (e.g. differences between urban and biomass

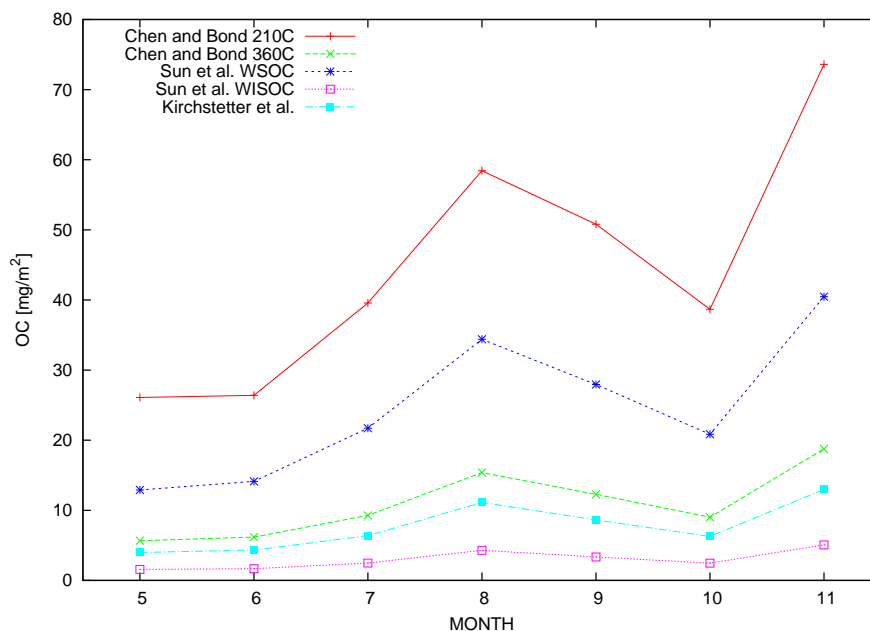


Fig. 4. Monthly mean absorbing OC [mg/m^2] for Mongu, assuming different imaginary index values for OC. Kirchstetter et al. (2004) based values correspond to the retrievals presented in this paper. Sensitivity study was carried out applying also imaginary index values presented in Chen and Bond (2010) for two production temperatures and in Sun et al. (2007) for water soluble (WSOC) and water-insoluble organic carbon (WISOC).

burning dominated sites) and temporal variability in light-absorbing organics are needed. Our study is a step towards this direction and offers the first AERONET-based estimates of absorbing organic carbon concentrations. Future work is needed on the refractive indexes of several absorbing OC species, to improve the understanding of abundance of absorbing OC.

Our main findings are:

(1) Absorbing OC retrievals from Beijing and XiangHe clearly dominates in winter season (up to $35 mg/m^2$), and they show highest values also in other seasons except for September–November, when retrievals from Kanpur and Gandhi College exhibit highest values due to very high biomass burning occurring in this season. In the summer the values of absorbing OC concentration are very similar in US and Europe (around $5 mg/m^2$), while in the Southern Hemisphere they are typically around $15\text{--}20 mg/m^2$.

(2) In any season, the grid-box including Beijing and XiangHe exhibits the highest OC/BC ratio, and the one including Kanpur and Gandhi College has also high OC/BC ratio. In the summer season the OC/BC ratios in Alaska and around Moscow were rather high. These OC and BC emissions are mostly from boreal forest fires.

(3) The substantial absorbing OC values we find at several sites are caused by enhanced aerosol absorption at shorter wavelengths. This is either due to absorbing OC compounds under assumption of constant refractive indexes for BC, due

to the remaining dust influence that we attempted to exclude or due to enhanced BC absorption at short wavelengths. In any case this enhanced aerosol absorption is very relevant for the radiative forcing of the direct aerosol effect.

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References

- Alexander, D. T. L., Crozier, P. A., and Anderson, J. R.: Brown carbon spheres in East Asian outflow and their optical properties, *Science*, 321, 833836, 2008.
- Andreae, M. O., and P. Merlet, Emission of Trace Gases and Aerosols From Biomass Burning, *Global Biogeochem. Cycles*, 15(4), 955–966, 2001.
- Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6, 3131–3148, 2006.
- Bergstrom, R. W., Pilewskie, P., Russell, P. B., Redemann, J., Bond, T. C., Quinn, P. K., and Sierau, B.: Spectral absorption properties of atmospheric aerosols, *Atmos. Chem. Phys.*, 7, 5937–5943, 2007.

- Bond, T., and R. Bergstrom, Light absorption by carbonaceous particles: An investigative review, *Aerosol Sci. Technol.*, 40(1), 27-67, 2006.
- Cao, J. J., et al., Spatial and seasonal distributions of carbonaceous aerosols over China, *J. Geophys. Res.*, 112, D22S11, doi:10.1029/2006JD008205, 2007.
- Chen, Y. and Bond, T. C.: Light absorption by organic carbon from wood combustion, *Atmos. Chem. Phys.*, 10, 1773-1787, doi:10.5194/acp-10-1773-2010, 2010.
- Dey, S., S. N. Tripathi, R. P. Singh, and B. Holben, Retrieval of black carbon and specific absorption over Kanpur city, Northern India during 2001-2003 using AERONET data, *Atmospheric Environment*, 40(3), 445-456, 2006.
- Dubovik, O., and M. D. King, A flexible inversion algorithm for retrieval of aerosol optical properties from Sun and sky radiance measurements, *J. Geophys. Res.*, 105(D16), 20,673-20,696, 2000.
- Eck, T. F., et al., Optical properties of boreal region biomass burning aerosols in central Alaska and seasonal variation of aerosol optical depth at an Arctic coastal site, *J. Geophys. Res.*, 114, D11201, doi:10.1029/2008JD010870, 2009.
- Ganguly, D., P. Ginoux, V. Ramaswamy, D. M. Winker, B. N. Holben, and S. N. Tripathi, Retrieving the composition and concentration of aerosols over the Indo-Gangetic basin using CALIOP and AERONET data, *Geophys. Res. Lett.*, 36, L13806, doi:10.1029/2009GL038315, 2009.
- Han, L., Zhuang G., Cheng, S., Li, J., The mineral aerosol and its impact on urban pollution aerosols over Beijing, China, *Atmospheric Environment*, doi:10.1016/j.atmosenv.2007.05.046, 2007.
- Holben B.N., T.F. Eck, I. Slutsker, D. Tanre, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan, Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, AERONET - A federated instrument network and data archive for aerosol characterization, *Rem. Sens. Environ.*, 66, 1-16, 1998.
- Ichoku, C., L. Giglio, M. J. Wooster, and L. A. Remer, Global characterization of biomass-burning patterns using satellite measurements of Radiative Energy, *Remote Sens. Environ.*, 112, 2950-2962, 2008..
- Intergovernmental Panel on Climate Change (IPCC), *Climate Change 2007: The Scientific Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., Cambridge Univ. Press, New York, 2007.
- Jacobson, M. Z., Isolating nitrated and aromatic aerosols and nitrated aromatic gases as sources of ultraviolet light absorption, *J. Geophys. Res.*, 104(D3), 3527-3542, doi:10.1029/1998JD100054, 1999.
- Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., and Wilson, J.: Organic aerosol and global climate modelling: a review, *Atmos. Chem. Phys.*, 5, 1053-1123, 2005.
- Kirchstetter, T. W., T. Novakov, and P. V. Hobbs, Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, *J. Geophys. Res.*, 109, D21208, doi:10.1029/2004JD004999, 2004.
- Koch, D., et al., Evaluation of black carbon estimations in global aerosol models, *Atmos. Chem. Phys. Discuss.*, 9, 15769-15825, 2009.
- Koven, C. D., and I. Fung, Inferring dust composition from wavelength-dependent absorption in Aerosol Robotic Network (AERONET) data, *J. Geophys. Res.*, 111, D14205, doi:10.1029/2005JD006678, 2006.
- Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, *Atmos. Chem. Phys.*, 10, 4207-4220, doi:10.5194/acp-10-4207-2010, 2010.
- Laskin J, A Laskin, PJ Roach, GW Slysz, GA Anderson, S Nizkorodov, DL Bones, and L Nguyen, High-Resolution Desorption Electrospray Ionization Mass Spectrometry for Chemical Characterization of Organic Aerosols. *Analytical Chemistry* 82(5):2048-2058. doi:10.1021/ac902801f, 2010.
- Lewis, K., W. P. Arnott, H. Moosmüller, and C. E. Wold, Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, *J. Geophys. Res.*, 113, D16203, doi:10.1029/2007JD009699, 2008.
- Lin, P., M. Hu, Z. Deng, J. Slanina, S. Han, Y. Kondo, N. Takegawa, Y. Miyazaki, Y. Zhao, and N. Sugimoto, Seasonal and diurnal variations of organic carbon in PM_{2.5} in Beijing and the estimation of secondary organic carbon, *J. Geophys. Res.*, 114, D00G11, doi:10.1029/2008JD010902, 2009.
- Luhar, A K., Mitchell, R M., Meyer, C P M., Qin, Y., Campbell, S K., Gras, J L., and Parry, D.: Biomass burning emissions over northern Australia constrained by aerosol measurements: II model validation, and impacts on air quality and radiative forcing, *Atmos. Environ.*, 42, 1647-1664, 2008.
- Martins, J. V., P. Artaxo, Y. J. Kaufman, A. D. Castanho, and L. A. Remer, Spectral absorption properties of aerosol particles from 350-2500nm, *Geophys. Res. Lett.*, 36, L13810, doi:10.1029/2009GL037435, 2009.
- Myhre, Gunnar, T.F. Bergelen, M. Johnsrud, C.R. Hoyle, Terje Berntsen, S.A. Christopher, D.W. Fahey, Ivar S. A. Isaksen, T.A. Jones, R.A. Kahn, N. Loeb, P. Quinn, L. Remer, J.P. Schwartz and K.E. Yttri, Modelled radiative forcing of the direct aerosol effect with multi-observation evaluation. *Atmos. Chem. Phys.*, 9: pp. 1365-1392, 2009.
- Myhre, G.: Consistency between satellite-derived and modeled estimates of the direct aerosol effect, *Science*, 325, 187-190, doi:10.1126/science.1174461, 2009.
- Qin, Y. and Mitchell, R. M.: Characterisation of episodic aerosol types over the Australian continent, *Atmos. Chem. Phys.*, 9, 1943-1956, doi:10.5194/acp-9-1943-2009, 2009.
- Ram, K., M. M. Sarin, S. N. Tripathi, One-year record of carbonaceous aerosols from urban site in the Indo- Gangetic Plain: Characterization, sources and temporal variability, under review, *JGR*, 2010.
- Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P.: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, *Atmos. Chem. Phys.*, 5, 799-825, 2005.
- Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos. Chem. Phys.*, 10, 1155-1169, doi:10.5194/acp-10-

- 1155-2010, 2010.
- Sato, Mki., J. Hansen, D. Koch, A. Lacis, R. Ruedy, O. Dubovik, B. Holben, M. Chin, and T. Novakov, Global atmospheric black carbon inferred from AERONET. *Proc. Natl. Acad. Sci.* 100, 6319-6324, 2003.
- Schuster, G. L., O. Dubovik, B. N. Holben, and E. E. Clothiaux, Inferring black carbon content and specific absorption from Aerosol Robotic Network (AERONET) aerosol retrievals, *J. Geophys. Res.*, 110, D10S17, doi:10.1029/2004JD004548, 2005.
- Schuster, G. L., B. Lin, and O. Dubovik, Remote sensing of aerosol water uptake, *Geophys. Res. Lett.*, 36, L03814, doi:10.1029/2008GL036576, 2009.
- Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Dunlea, E. J., Roberts, G. C., Tomlinson, J. M., Collins, D. R., Howell, S. G., Kapustin, V. N., McNaughton, C. S., and Zhou, J.: Aerosol optical properties relevant to regional remote sensing of CCN activity and links to their organic mass fraction: airborne observations over Central Mexico and the US West Coast during MILAGRO/INTEX-B, *Atmos. Chem. Phys.*, 9, 6727-6742, doi:10.5194/acp-9-6727-2009, 2009.
- Sun, H., L. Biedermann, and T. C. Bond, Color of brown carbon: A model for ultraviolet and visible light absorption by organic carbon aerosol, *Geophys. Res. Lett.*, 34, L17813, doi:10.1029/2007GL029797, 2007.
- Tripathi, S. N., S. Dey, V. Tare, and S. K. Satheesh: Aerosol black carbon radiative forcing at an industrial city in northern India, *Geophysical Research Letters*, 32, doi:10.1029/2005GL022515, 2005a.
- Tripathi, S. N., S. Dey, V. Tare, S. K. Satheesh, S. Lal and S. Venkataramni:, Enhanced layer of black carbon in a north Indian industrial city, *Geophys. Res. Lett.*, 32(12), L12802, doi:10.1029/2005GL022564, 2005b.
- Turpin B.J. and H.J. Lim , Species contributions to PM_{2.5} mass concentrations: Revisiting common assumptions for estimating organic mass. *Aerosol Science and Technology* 35, pp. 602-610, 2001.
- Venkataraman C., G. Habib, A. Eiguren-Fernandez, A.H. Miguel and S.K. Friedlander, Residential biofuels in South Asia: Carbonaceous aerosol emissions and climate impacts, *Science*, 307(5714), 1424-1426, 2005.
- Yang, M., Howell, S. G., Zhuang, J., and Huebert, B. J.: Attribution of aerosol light absorption to black carbon, brown carbon, and dust in China – interpretations of atmospheric measurements during EAST-AIRE, *Atmos. Chem. Phys.*, 9, 2035-2050, 2009.

Table 1. Refractive index values of the components, real part (n) and imaginary part (k). All the components are assumed to be spectrally constant, except for OC whose imaginary index at 440nm and 670nm is by Kirchstetter et al. (2004) and zero at 870nm and 1020nm. Shown are also the imaginary index values for OC, based on Chen and Bond (2010) and Sun et al. (2007), that were used to assess the sensitivity of the OC retrieval for the assumed imaginary index.

	n	k
Water	1.33	1.96×10^{-9}
$(NH_4)_2(SO_4)$	1.53	10^{-7}
Soot	1.95	0.79
OC, Kirchstetter et al. (2004)	1.53	0.073/0.0034 (440nm/670nm)
OC, Chen and Bond (2010), 210C	1.53	0.0122/0.0013 (440nm/670nm)
OC, Chen and Bond (2010), 360C	1.53	0.0525/0.0050 (440nm/670nm)
OC, Sun et al. (2007), WSOC	1.53	0.0232/0.0032 (440nm/670nm)
OC, Sun et al. (2007), WISOC	1.53	0.1908/0.0131 (440nm/670nm)

Table 2. Comparison of monthly mean OC (mg/m^2) from Oslo CTM and those retrieved from AERONET imaginary index. First are the AERONET-based OC concentrations (fine mode only in parenthesis), the model-based OC concentrations after the slash.

Month	Beijing	Kanpur	Mongu	Alta Floresta	Gandhi College
1	38.29 (21.92) / 5.84	16.29 (9.95) / 7.45	— (—) / -99.00	— (—) / 8.87	— (—) / 10.07
2	33.20 (19.58) / 9.20	16.84 (10.27) / 8.45	— (—) / -99.00	— (—) / -99.00	— (—) / 9.66
3	28.28 (16.54) / 3.85	— (—) / 10.35	— (—) / -99.00	— (—) / -99.00	— (—) / 10.55
4	27.81 (16.25) / 7.55	— (—) / 6.69	— (—) / -99.00	— (—) / -99.00	— (—) / 8.78
5	20.86 (11.79) / 6.49	— (—) / 5.54	5.97 (4.08) / -99.00	— (—) / 15.87	— (—) / 8.63
6	20.22 (11.94) / 9.35	— (—) / 6.99	6.50 (4.70) / 19.18	— (—) / 24.58	— (—) / 8.92
7	12.74 (8.49) / 11.85	— (—) / 8.57	9.56 (6.39) / 22.30	— (—) / 36.60	— (—) / 8.31
8	19.83 (11.78) / 11.88	— (—) / 9.62	16.70 (11.37) / 28.04	17.65 (13.82) / 93.39	— (—) / 9.45
9	22.62 (13.04) / 9.90	11.04 (7.17) / 15.13	12.94 (8.57) / 26.74	19.86 (15.55) / 55.90	— (—) / 17.03
10	29.20 (16.43) / 8.72	32.71 (17.81) / 14.60	9.40 (6.12) / 19.21	7.94 (5.73) / 30.92	12.36 (6.98) / 18.32
11	41.91 (23.56) / 7.86	36.74 (20.43) / 10.08	19.46 (12.57) / 14.74	— (—) / 13.93	41.86 (24.68) / 12.42
12	51.10 (28.85) / 7.31	20.75 (11.94) / 10.97	— (—) / -99.00	— (—) / 9.97	30.30 (20.64) / 11.68