EMEP/ACTRIS intensive measurement period – Winter 2018

Wenche Aas, Stephen Platt and Karl Espen Yttri (EMEP – CCC, NILU)

Background

Carbonaceous aerosol is a major fraction of the ambient aerosol in Europe. It influences the atmospheric radiative balance and contributes to adverse health effects. Consequently, carbonaceous aerosol is a key species measured regularly in air quality networks, such as EMEP.

Carbonaceous aerosol sources are abundant and both anthropogenic and natural, thus identifying and quantifying sources are important to develop efficient abatement strategies. In particular, there is an interest to distinguish the relative contribution of combustion sources making use of either fossil fuels (such as road transportation), or biomass (such as residential wood burning). Source apportionment (SA) of the carbonaceous aerosol is usually based on filter samples with a 24-hour time resolution or more, and typically includes several species that are not part of regular monitoring. Detailed separation of sources is challenging and a certain overlap between apportioned sources is likely. Further, the various SA approaches are costly, and results are only available after sample collection.

Consequently, there is a demand for high time resolution and on-line information on aerosol particles from specific sources, not only from scientists wanting to understand atmospheric dynamics and composition, but also for air-quality managers to inform, educate and increase public awareness of air-quality related issues and to design air quality management plans. On-line and high time-resolution measurements by e.g. aerosol mass spectrometers have become available in recent years. However, the mass spectra must be processed post sampling to obtain source-apportioned data of the carbonaceous aerosol, and are thus not available on-line. Separation of equivalent black carbon (EBC) into fossil fuel (EBC_{ff}) and wood burning (EBC_{wb}) sources is possible by multi wavelength measurement of the absorption coefficient (Sandradewi et al., 2008), and for the AE33 aethalometer this is an online feature.

A few European studies have reported using the multi wavelength aethalometer for source apportionment of EBC, and even in the rural background environment, and for an extended period of time (Herich et al., 2011; Zotter, et al. 2017; Martinsson et al. 2017). With an increasing number of multi wavelength aethalometers employed at European rural background sites, as part of regular monitoring, and the substantial focus on BC and its sources (wood burning in particular), it appears timely to focus on such measurements in a forthcoming winter time EMEP/ACTRIS intensive measurement period (IMP).

The IMP should aim to test the multiwavelength aethalometer source apportionment approach in the European rural background environment, including low loading areas in Scandinavia and more polluted regions in Central Europe, and in areas differing in source composition, preferably also with an influence of coal combustion. Further, it should compare EBC_{bb} and EBC_{ff} apportioned by the multiwavelength aethalometer approach to filter based measurements of the biomass burning tracer levoglucosan and elemental carbon (EC) for validation purposes.

Choice of Absorption Angström Exponent

The multi wavelength aethalometer approach for separating EBC into EBC_{bb} and EBC_{ff} is based on the assumption that aerosol particles emitted from wood burning absorbs relatively more in the near UV than in the IR, compared to aerosol particles from combustion of fossil fuels, which show no wavelength dependence.

The choice of the absorption Angström exponent (AAE) is decisive for the separation of EBC into EBC_{bb} and EBC_{ff}. Wood burning emissions are assumed to have an AAE ranging from 1.5 to 2.5, whereas it is about 1 for fossil fuel emissions. Firstly, we will apportion EBC_{bb} and EBC_{ff} by using positive matrix factorisation and ME2 via the SoFi toolkit (Canonaco, 2013). Selecting a two-factor solution, we take the log natural of the absorption and constrain the slope of the wavelength dependence of one factor to 1.0±0.25 (representing fossil fuel combustion) and the other to 2.0±0.5 (representing biomass burning). In this way, we emulate the Sandradewi et al. (2008) approach, but use the more powerful PMF solver to apportion the sources without any strong a priori assumptions on the AAE. The PMF output is evaluated via comparison of the biomass burning factor to levoglucosan (see "Validation" Section). The slope of the wavelength dependence of each factor yields a site specific AAE for fossil fuel combustion and biomass burning. This approach ensures an optimized solution for each of the participating sites and ought to be superior to choosing one factor for each of the two sources to be valid for all of Europe because of the different typology of fuel used throughout Europe.

Complementary to the PMF-approach, we will use the optimisation technique of Fuller et al. (2014): By systematically varying the AAE used for EBC_{ff} in the Sandradewi et al. (2008) approach, we will select that AAE for which the regression intercept of the levoglucosan vs. EBC_{bb} is zero.

Validation

Validation of the source apportioned EBC_{bb} and EBC_{ff} from the multiwavelength aethalometer approach is essential, and is typically performed using levoglucosan and/or fossil fuel (ff) and biomass burning (bb) TC/OC/EC, derived from ^{14}C analysis (potassium and NO_x have also been used).

For a winter (December, January, February) sampling period, atmospheric degradation of levoglucosan should be considered minor, and thus highly suitable for validation purposes, although a certain influence of levoglucosan from combustion of brown coal cannot be excluded for certain parts of Europe. TC_{bb} and OC_{bb} derived from ^{14}C-analysis can be influenced by other sources of modern carbon (e.g. biogenic secondary organic aerosol (BSOA) and primary biological aerosol particles (PBAP)), although less pronounced in winter, whereas EC_{bb} from ^{14}C-analysis is not prone to this, although the separation of EC and OC can add some uncertainty. EC_{ff} and EC_{bb} derived from ^{14}C-analysis can be compared to aethalometer estimates of EBC_{ff} and EBC_{bb}, whereas levoglucosan is a tracer for EBC_{bb} only. Combining levogluosan with concurrent EC measurements and a (levoglucosan/EC_{bb}) ratio, provides an estimate of EC_{ff}, but with larger uncertainty than EC_{ff} derived from ^{14}C-analysis.

Validation of the EBC_{bb} and EBC_{ff} by levoglucosan/^{14}C-derived EBC_{ff}/EBC_{bb}, is done by correlation, assuming that a high correlation between e.g. EBC_{bb} and levoglucosan means that the wood burning signal from the aethalometer is well reproduced; i.e. a qualitative validation. A quantitative comparison of different approaches is possible, e.g. by comparing EBC_{bb} from the aethalometer approach with EC_{bb}, obtained from observed levels of levoglucosan (EC_{bb} = [levo] x $(EC/levo)_{bb}$).

There are a large number of laboratories conducting levoglucosan analysis, and with a rather good comparability, there is no need for a centralized laboratory, as is the case for ^{14}C-analysis. Levoglucosan analysis is also noticeably less expensive than ^{14}C-analysis, allowing for analysis of a larger number of samples. Hence, we suggest that levoglucosan is analyzed from quartz fiber filters from a co-located sampler, along with EC, OC and TC.

With concurrent measurements of EBC_{bb}, EBC_{ff}, and TC, it is possible to calculate the carbonaceous aerosol from fossil fuel and wood burning, as well that of a non-light absorbing fraction. Attempts on this are reported in the scientific literature, but with various degrees of success. Conducting the IMP in winter could increase the possibility of a successful outcome, but it is our opinion that it should not be a major aim of the IMP.

Participation, partnership and co-benefit

All EMEP/ACTRIS sites performing absorption coefficient measurements with a multi-wavelength aethalometer (AE31 or AE33), are invited to participate in the proposed EMEP/ACTRIS intensive measurement period.

The proposed initiative has a thematically strong connection to ongoing activities within ACTRIS, and has the possibility to benefit from these. Apportionment of EBC_{bb} and EBC_{ff} by multi-wavelength measurements of the absorption coefficient is however not a specific task of ACTRIS, although the infrastructure is already there: Calibrated multi wavelength instruments is available for quite a few sites, as is inter compared analytical methods for levoglucosan and OC/EC/TC, due to the European Center for Aerosol Calibration (ECAC) activity.

The proposed initiative will initiate measurements of levoglucosan (at least for a period of time), which is a deliverable in ACTRIS, as well as high time resolution measurements of the wood burning source (EBC_{bb}). Further, it will provide high time resolution measurements of EBC_{ff}, a tracer of anthropogenic combustion of fossil fuel, likely to encompass, but not exclusively, the sources otherwise accounted for by hopanes/steranes and nitro-PAHs, listed by ACTRIS as being of particular interest, but less likely to be implemented in regular monitoring than e.g. levoglucosan. The topic proposed for the forthcoming IMP is in line with the EMEP monitoring strategy.

A successfully initiated IMP would likely greatly benefit from a cooperation with the recently established COST action COLOSSAL (Chemical On-Line cOmpoSition and Source Apportionment of fine aerosol).

Experimental
Multi wavelength measurement of the absorption coefficient by an aethalometer (AE31/AE33) and a co-located filter sampler with a size segregating inlet with a cut-off size matching that of the aethalometer, is a prerequisite for participation in the IMP. The collected quartz fibre filters should be analysed with respect to levoglucosan, EC, OC and TC, as a minimum. The filter sampler should be the same sampler commonly used for monitoring of OC/EC/TC at the site, if possible. The time resolution of the aethalometer and the filter sampler should be identical to that of regular monitoring at the premises. This is to be sure that filter loading is adapted to the ambient level experienced at the actual site, avoiding too low or too high loading, and because the validation ought to be based on the procedures already used for the long term monitoring at the site.

The number of samples available for EC, OC, TC and levoglucosan analysis, and thus for validation of the aethalometer approach, depends on the sampling time and the frequency of filter collection by the co-located filter sampler. 25 – 30 filter samples ought to be analysed for validation purpose, which is possible to obtain for all sites with a sampling time of 72 hours or less and with continuous measurements. For sites with a discontinuous sampling regime, an increased frequency during the IMP is a possibility. Low loading sites with a weekly sampling frequency may need to expand the sampling period to obtain a sufficient number of samples.

Documented calibration of the aethalometer at ECAC is warranted, but lack of such is not disqualifying. OC, EC, and TC should be analysed according to the EUSAAR-2 protocol, and participation in a recent or upcoming ILC is highly encouraged. Levoglucosan, as for OC, EC, and TC, can be analysed by own laboratory facilities, preferably by a method that has been intercompared (e.g. by Yttri et al., 2015), or that in another way has documented to be compliant.

Participants not performing OC, EC, TC or levoglucosan analysis, will have the possibility to get their samples analysed at a centralized laboratory at cost price.

Additional analysis of relevant species (e.g. PAHs, picene) and by instruments such as ACSM are encouraged, but not requested. To further link the black carbon mass to an absorption coefficient it would be beneficial if some sites could run a MAAP instrument in parallel. (See presentation by Nicolas Bukowiecki at the TFMM workshop in 2016: http://www.nilu.no/projects/ccc/tfmm/utrecht_2016/pres/ACTRIS-2_JRA1_and_EMEP_MAC_ap.pptx)

The IMP is scheduled for winter 2018, and could start as early as December 2017. It should take place for a period of at least three months and which coincides with the time period experiencing the highest concentrations of EBC. Measurements should take place during the same time period at all sites.

Summary

Aim

- To quantify EBC_{ff} and EBC_{bb} by multi wavelength aethalometer measurement, and to validate this approach using concurrent off-line measurements of the wood burning tracer levoglucosan (and EC, OC, TC) for a wider part of Europe.
- To provide a harmonized data set for model validation
- To initiate regular monitoring of EBC_{ff} and EBC_{bb}, and reporting of such data to EBAS.

Time schedule

- The IMP is scheduled for winter 2018, and could start as early as December 2017.
- To be proposed jointly to EMEP Steering Body in September and at the ACTRIS technical meeting decision in October. Coordinator for the practical part of the campaign will be at CCC, scientific lead to be decided later, most likely at the ACTRIS October meeting.

Costs

- The participants will finance their own running costs.

Participants not performing OC, EC, TC or levoglucosan analysis, will have the possibility to get their samples analysed at a centralized laboratory at cost price.: Estimated costs for TOA analysis (EUSAAR-2) of OC, EC and TC: 70€ pr. sample (ex. VAT) and for levoglucosan analysis: 100 € pr. sample (ex. VAT).

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