Summary

The BelEUROS model has been optimized in the framework of this study with respect to its capacity to calculate the chemical composition of PM_{10}, i.e. the chemical subcomponents of fine particulate matter. This optimization has been carried out by a comparison of model results with the observations collected during the Chemkar II and Chemkar III field campaigns of VMM. The optimized model version has been validated subsequently.

The main results of this validation exercise are that BelEUROS on the one hand succeeds to calculate the secondary inorganic compounds (ammonium, nitrate and sulfate) quite correctly, especially the annual average concentrations, and the results obtained are sufficiently reliable to use them in studies. On the other hand, it is currently not possible to calculate the concentrations of the primary particulate components (elemental carbon (EC), organic carbon (OC) and mineral dust) in a sufficiently reliable way.

For EC, we find, depending on the location and the time period, more or less significant overestimates of the observed concentrations and for OC and mineral dust BelEUROS underestimates the observed concentrations clearly in general. For the sum of the primary particulate components (PPM_{10}) typically the low concentrations are overestimated by the model and the high concentrations are underestimated. Also, differences in model performance have been detected between the Chemkar II- and the Chemkar III-field campaigns. Observed correlation coefficients are higher for the period of the Chemkar II-campaign for most components.

Several options have been tested to calibrate the model for the sum of the primary particulate components (PPM_{10}). A calibration based on the obtained correlation between measured and modeled PPM_{10}-concentrations yielded no satisfactory result because the model underestimates the variation in measured PPM_{10}-concentrations strongly. Hence, this calibration method leads to a far too large variation in calculated PPM_{10}-concentrations, including some negative daily average concentrations. This calibration method is potentially suitable for the calculation of an annual average concentration, but not for the calculation of daily average concentrations, as required in this study.

A possible alternative calibration method, which was also tested, is based on the addition of the concentration-dependent bias between the modeled and observed PPM_{10}-concentration. This method would make much greater use of the PPM_{10}- or PM_{10}-measurements for the considered location and for the same day. This method gives basically good results regarding the comparison of measurement and model values. However, two important questions arise regarding this method: First, does the model actually offer an added value when using this calibration technique compared to the use of an estimate of PPM_{10} purely based on the PM_{10} observations (or PM_{10}-concentrations from the RIO-model)? Secondly, might there not be an artifact when using this calibration method because on days with high PM_{10}-concentrations a much larger bias (a much larger PPM_{10}-concentration) is added than on days with low PM_{10}-concentrations? This would have an impact on the calculated concentrations of PPM_{10} on exceedance days of the PM_{10} daily limit value compared to the annual average concentration of PPM_{10}. The advantages and disadvantages of the different calibration methods were investigated.

The conclusions of this exercise can be summarized as follows:

The results of the model validation indicate that the modeling of individual primary particle components is still insufficiently correct to draw sound conclusions about sources and causes of exceedance days. There are basically four options for the model to be operated:

1. without calibration;
2. with a simple calibration based on y = ax + b;
3. with a bias correction for PPM_{10} based on daily measurements, and, finally;
4. with a replacement of the modeled PPM_{10} by an estimate based on the PM_{10} (or PM_{10} concentrations from RIO).

The advantages and disadvantages of these 4 options are summarized in the following table:

Optie	Advantage	Disadvantage
1	No possible artifact by the bias-correction due to stronger correction on exceedance days than on non-exceedance days.	PPM_{10} is clearly underestimated, as shown in the model validation. With this option, no realistic PPM_{10} concentrations are calculated.
2	No possible artifact by the bias correction, the average PPM_{10}-concentration corresponds to the one of the measurements.	Very high scatter on the calculated results, including many negative concentrations; this method does not seem suitable for use on daily average concentrations.
3	Good agreement between model and measurements, even at high concentrations of PPM_{10}.	Danger exists that due to the concentration-dependent bias correction an artifact arises because much stronger correction is applied on exceedance days than on non-exceedance days.
4	Use of (PM_{10}) measurements with known uncertainty.	Because of few measurements on exceedance days in Chemkar II the correlation between measured PM_{10} and PPM_{10} is possibly not representative for exceedance days; transfer of the results to other locations and times is difficult.

The fundamental problem regarding option 2, which is normally preferred for the calibration of models, is that the scatter of the obtained results is very large. This means that the model is not suitable for the calculation of PPM_{10} on a daily basis. The reason behind this is probably connected to a (large) part to the absence and/or underestimation of certain, probably mainly Flemish, emission data, especially (diffuse) primary particle emissions from e.g. residential wood combustion, resuspended dust from road traffic and dust emissions from storage and handling of bulk goods.

The most realistic results can be obtained with options 3 and 4. These seem to be the best way to address the issue of the composition on exceedance days. Essentially, options 3 and 4 mean that PPM_{10} is estimated more accurately by the use of measurements. Options 1 and 2 are based on the model (whether or not calibrated in the classical way), options 3 and 4 are a combination of model and measurements.

In brief, the model confirms more or less the measurements of secondary inorganic PM_{10} components but it is not yet suitable for PPM_{10} and we must therefore conclude with the information we get from the measurements, especially regarding the contribution of PPM_{10} on exceedance days.

The latter however is also problematic to some extent because during the Chemkar II measurement campaign too little exceedance days were included in the measurements of the chemical composition. Hence, no decision can be made about the chemical composition of PM_{10} on exceedance days compared to the (annual) average chemical composition. E.g., in Aarschot only 2 out of the 31 exceedance days during the Chemkar II measurements of the chemical composition were included. Also on other measurement locations this rate is very low. For this reason we have to conclude that we currently (for the Chemkar II campaign for which the model calibration was set up) cannot make a reliable statement about the contribution of the chemical subcomponents of PM_{10} on exceedance days compared to the annual average contributions, neither based on model results nor based on measurements.

In the last part of the study, the Flemish contribution has been calculated by chemical subcomponent and also here we tried to distinguish between the annual average Flemish contribution and the Flemish contribution on exceedance days.

Regardless of all the above mentioned uncertainties associated with the modeling results there are a number of conclusions that can be drawn from these calculations with a sufficient reliability. The calculations have shown that the Flemish contribution to the concentrations of EC, OC and the mineral fraction is significantly higher than the Flemish contribution to the total PM_{10}-fraction. With values of up to 80 or 90 % Flemish contribution to the EC-concentrations in Borgerhout it is very clear that this chemical component is for a very large part derived from Flemish sources. Additionally, currently the adverse health effects associated with PM_{10} are to a large part allocated to EC (e.g. Janssen et al., 2011). If we compare this value with the calculated contribution of Flemish emissions to PM_{10}-concentrations of about 25 % on average over Flanders a completely different picture for EC (and other primary components) is seen. Generally, we find significantly higher contributions for the PPM_{10}-components from Flemish sources than for the secondary PM_{10}-components. At that moment it is assumed that larger health hazards are associated with the primary particulate components than with the secondary particulate components, hence this result corrects somewhat the impression that might arise from the number of 25 % mean Flemish contribution to PM_{10}-concentrations.

The health relevant fractions (mainly EC and OC) of PM_{10} thus exhibit high Flemish contributions to their concentrations, and therefore need to be addressed on the Flemish policy level. Measures taken abroad will only have a relatively small effect on the concentrations of these pollutants in Flanders. Opportunities for a decrease of the EC-concentrations as a result of local measures include low-emission zones, road pricing and other measures to reduce the amount of traffic. The retrofit of existing diesel engines, for which a subsidy system exists in Flanders, may also contribute.

Furthermore, it is clear that the Flemish contribution to primary PM_{10} and to total PM_{10} is significantly higher in urban areas than on average in Flanders. This result is related to the origin of the emissions that contribute to the exposure of large parts of the population.

Concerning the uncertainties on these results, in addition to the above discussed uncertainties of the model itself and possible calibration uncertainties, additional important uncertainties are related to the used emissions. There are many indications that a number of local contributions, especially of primary particulate matter emissions (e.g. by wood combustion and resuspended road dust), are not or only partially included in the emission inventory. This means we have to assume that the Flemish contribution to PM_{10} is probably underestimated by the model because these (missing) local particulate emissions would lead to a higher contribution of local sources if included into the model. It is important to note that BelEUROS (and many other similar models) only estimate the emissions of a number of natural sources itself, i.e. the emissions of sea salt and of isoprene and terpenes by vegetation. The models themselves do not estimate any anthropogenic emissions. If these emissions are not used in the emission inventory or are underestimated the model will inevitably miss or underestimate the contributions from these sources.

This would also have an effect on the calculation of e.g. the contributions of the various socio-economic sectors in Flanders to the PM_{10}-concentrations. For example, suppose the primary particulate emissions from one sector are quite correctly estimated in the emission inventory (due to the fact that there were already many studies conducted on this subject in Flanders), those of another sector are instead underestimated, this would result in a distorted picture when calculating sector contributions purely because of the different degree of completeness of the primary dust emission inventory for the different sectors.

We can thus conclude that the highest priority should currently be given to the completion of the emission inventory for primarily particulate matter. Once these emissions are better included, the model performance most likely will (automatically) improve. This seems at the moment even more important than further improvements of the model itself. As the model is concerned, the search for better calibration methods for PPM_{10} is especially important as this point is currently one of the main problems. From this both retrospective calculations but also the PM_{10}-predictions would benefit.