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Interactive comment on “Spatial variations and development of land use regression models of levoglucosan in four European study areas” by A. Jedynska et al.

Anonymous Referee #1

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The majority of this paper describes a very limited dataset of levoglucosan measurements made in four regions, with a rather minor part of the paper describing the development of a land use regression model. The main weakness of the paper is the extremely small dataset upon which it is based. This comprises just three 2-weekly samples collected over a period of one year at each of the sites. These were taken in winter, summer and an intermediate season (either spring or autumn). Correlations with other pollutants that were measured more intensively were used to estimate an annual mean from this very small dataset, but no attempt is made to estimate the uncertainties in that annual mean. It appears that correction of the annual mean was

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based on temporal correlations for each site between levoglucosan and other pollutants such as NO_x based upon the three samples. Whilst mathematically it may be possible to calculate a regression from three samples, it would not seem to be a very meaningful number. The existence of such temporal correlations seems quite surprising particularly as at three sites the pollutant to give the best temporal correlation with levoglucosan was NO_x which did not show spatial correlation with levoglucosan and does not vary in source strength appreciably through the year (much of the ground level NO_x comes from road traffic) while the levoglucosan source strength varies hugely with season. One is therefore left to wonder how robust the data are. Even the fact that no winter samples were collected in the Munich/Augsburg area did not apparently stop the authors going ahead with developing a land use regression model, despite the fact that winter to summer ratios were very large in this work.

There are also inconsistent arguments within the paper. Modest contrasts in levoglucosan concentrations across Europe are attributed to forest fires and agricultural burning being more common in southern Europe, whereas wood combustion for domestic heating etc. is more common in northern Europe. However, the data in this paper, while showing a rather modest spatial contrast in levoglucosan, show a huge seasonal variation hence suggesting that the contribution of forest fires and agricultural burning (which are summer phenomena) has not influenced the dataset.

The development of the land use regression model is described in very modest detail and it appears that most of the explanatory variables used initially related to road traffic and unsurprisingly were not predictive. The main outcomes of the land use regression model work appear in Table 5 and it is left for the reader to guess what time average the models refer to. Presumably, it is an annual mean, and if so, this needs to be stated. The fact that the models all use different predictive variables gives rather little confidence in the outcomes and this matter is not discussed. It may be that the four areas are so different that the controls on concentration are very different in nature, but there is minimal discussion of the meaning of these models and one has to take them

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on trust. The main reference in the test is to the r^2 values, which are apparently quite high, but the poorly explained LOOCV r^2 values, which apparently are used to indicate possible spatial auto-correlation in the residuals (not commented on subsequently) are much lower and suggest that the models for three of the four regions are explaining less than 50

Another major weakness is that no independent data were used to contribute to model validation (perhaps from a different year or from different two week periods within the same year) which if available would have given a valuable test of the results.

The *levo*/PAH ratios in Table S6 require explanation as they appear to imply PAH concentrations around 1-2 ng m⁻³ which appears low for total PAH.

Overall, this is an extremely marginal piece of work that does little to advance the science and needs substantial enhancement to justify publication.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 13491, 2014.

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