

We thank the reviewers (Dr. Mick Whelan and the anonymous reviewer #2) very much for their positive feedback and their valuable comments. We have tried to clarify what was unclear and addressed each of the comments below. All reviewers' comments are listed in this document in black and our answers to them are given below in blue and italic. We would be happy to include the suggested changes in a revised version of the manuscript.

M. Whelan (mjw72@le.ac.uk)

Received and published: 29 September 2013

General comments

This paper is concerned with assessing the importance of uncertainty in pesticide leaching model parameters relative to uncertainties in climate change predictions (as defined by a range of future climate predictions produced by different GCMs and emission scenarios). Overall, the paper is well written and of a high technical standard. The authors have made a good attempt at performing and presenting a thorough statistical analysis of their data and the findings presented will be interesting for researchers in both the climate change and pesticide leaching communities. The authors argue that whilst the uncertainties of CC have been explored extensively in many aspects of catchment hydrology, they have not yet been addressed for pesticide leaching. I think that this provides a reasonable rationale for the work presented. Provided that the authors can provide adequate responses to some specific questions and observations below, I would have no objection to this paper being published in HESS.

Specific Comments

Abstract: Line 1: risk OF pesticide leaching. My understanding of risk is that it is, strictly speaking, a function of both the likelihood of occurrence and the consequences of a potentially harmful event. Here, we are only concerned with likelihood – effects are not considered. I am aware that risk is used in the same way elsewhere in the literature but I would prefer to see it reserved for studies of true risk. Same applies to Line 24.

We understand the reviewer's concern and will skip the wording "risk" as we do not want to talk about risk assessment and our more general use of the word might be misinterpreted.

P 10463 Effects of temperature. Probably the biggest effect of increased temperature would be to increase PET. Whether this is manifested as an increase in AET will depend on the pattern of soil moisture content and any influence of a CO₂ fertilisation effect, which will probably increase transpirative efficiency. Comment?

Yes, it is true that we did not discuss in the paper the effect of temperature on potential evapotranspiration (PET). In our simulations, PET and AET both increase in the future, but the ratio between AET and PET decreased slightly compared to present conditions. As the reviewer wrote, AET also depends on the influence of CO₂-fertilization effect, which was not accounted for in our simulations. We also did not consider the fact that higher temperatures will probably induce faster crop development and thus, earlier harvest of crops, which could also influence the water balance. However, any effects on pesticide leaching are likely to be very small compared to the effects of changes in precipitation on the water balance and the

effects of increased temperatures on degradation. Nevertheless, we will add a sentence in the introduction regarding the additional effects of temperature on PET, AET, soil moisture, and water balance in general.

The idea of increased diffusion rates reducing preferential flow is interesting but diffusion is slow relative to advection so how much difference would even a big increase in D make? Presumably this is covered by Jarvis (1998)?

Yes, we are referring to equilibration of pesticide concentrations between matrix and macropores laterally in the soil due to diffusion, which takes place over small distances. Diffusion is then surprisingly fast and not slow relative to convection. This process is described by the model we used (MACRO).

I agree that a change in temperature should change the sorption of many pesticides. It is stated on p10466 that sorption was assumed to be temperature-dependent. However, I think some brief details of how this effect was accounted for in MACRO are warranted here (despite the fact that it may be covered in Steffens et al., 2013).

We will add some brief details and a further reference in a revised version to how we modeled this.

It is stated on p 10476 L 20, that the effects of temperature are larger for more strongly sorbing pesticides. This implies that these compounds have a higher (absolute) enthalpy of phase change. Was this accounted for here?

No, we assumed the same sorption enthalpy for all three hypothetical compounds. We will add this information in the paper.

If not, then this statement is speculative and should be modified accordingly to make clear that this was not included in the analysis. Surely, if the same enthalpy of phase change is assumed for all the hypothetical compounds then the slope of log KOC versus temperature should be the same and the change in sorption for a given change in temperature should also be the same? Comment required please.

Yes, it is true that the change in sorption for a given temperature change is the same for the three hypothetical compounds. We realized that our statements in connection to Fig. 5 were a little misleading and we will modify this text in a revised version.

P10466 Good to see that internal state variables were used in calibration and validation.

Yes, we considered it important to use both internal state variables and flux variables for calibrating the model (i.e. water content and drainflow, as well as resident concentrations of bentazone and bromide in addition to bentazone and bromide concentrations in the drainflow).

P10467 In principle, it is good that only two system parameters and two pesticide specific parameters were chosen for the uncertainty analysis as it avoids “over-fitting”. However, more justification is perhaps warranted here about why these parameters were “considered” uncertain.

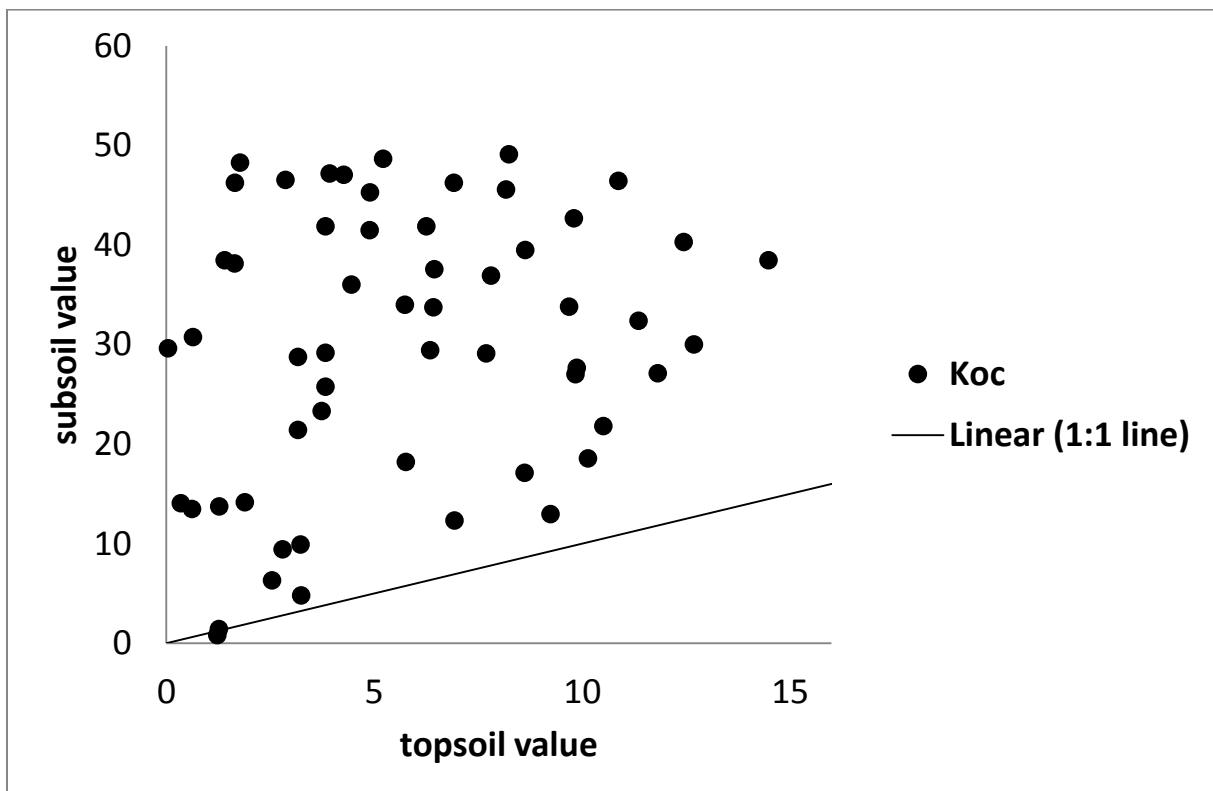
The two pesticide-specific parameters have been shown to be sensitive and uncertain in different pesticide leaching models (Boesten 1991; Dubus et al. 2003), and the two system parameters are sensitive for macropore flow (Dubus and Brown 2002, Larsbo and Jarvis 2005) and therefore considered in this study, which explicitly accounts for macropore flow (Larsbo et al. 2005). We will rephrase the text to clarify this and add relevant references on p 10467.

Why were uniform distributions assumed? This implies that all values in the range are equally likely. Is that really the case?

Uniform distributions as prior distribution are commonly used within GLUE-approaches (Beven and Binley, 1992; Liu et al. 2009; Nylander et al. 2011) as it reduces the dependency on the prior-guess if enough samples are taken. It is of course rather unlikely that the whole range of parameters has the same likelihood, but we did not have a better estimate of the prior distributions and wanted to test the whole range.

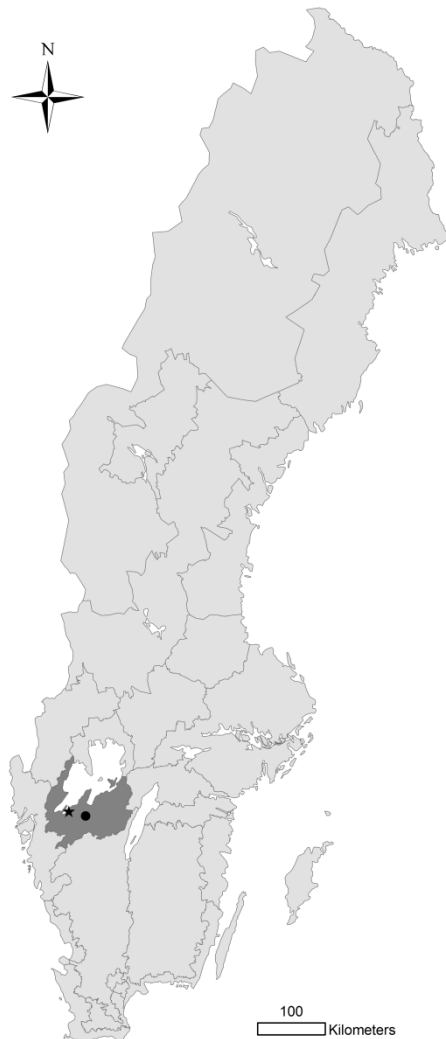
Also, why use a different KOC for topsoil and subsoil? Surely sorption will be normalized by the OC content of the soil layer. If KOC(top) and KOC(sub) are not correlated in the LHS, you could get simulations in which a very high value is chosen for one and a very low value for the other (or vice versa). Is this realistic? This comment is also relevant to the results and discussion presented on p10473.

There is very good reason to expect higher (effective) Koc values in the subsoil because inorganic soil constituents play a much more important role in sorption when organic carbon content is low (see e.g. Ghafoor et al. 2013), as it is in the Lanna subsoil. Instead of treating Koc values independently for topsoil and subsoil, we could have calibrated the ratio between those values, but we wanted to give the procedure the most freedom to choose the best fit itself, which should favor simulations that show certain correlations if they exist. As you can see from the graph below, the chosen subsoil Koc values were indeed always (with one exception) higher in the subsoil than in the topsoil and therefore rather consistent with our expectation. We will comment briefly on this in the revised paper.



P10468 How far is Lanna from the weather station at Sätenäs. Precipitation is extremely sensitive to spatial variations. I don't think this will have any major implications for the work described here but perhaps the authors might like to reassure the reader that this is the case?

The referee may have misunderstood this. We used on-site measurements of precipitation and other meteorological data for the model calibration (see p 10467, l 17/18; black dot in the figure below). We only used 30yr measured data from a weather station close by (Såtenäs; black star in the figure below) for the long-term scenario simulations. We considered this to be representative climate data for the region around Lanna.



P10472 Why was the reference climate disregarded?

We realized that the word “disregarded” is a little misleading and obviously confuses the reader. We will rephrase and clarify this in a revised version.

P10473 Fig 3. Why not show the frequency as a relative frequency? This would make more sense than counts.

Since the total number of different parameter values is always the same in all graphs (i.e.56), we did not consider it as important, but we could change it in a revised version, if necessary.

L13-14 re the inability of the model to simulate the 1st peak. It looks to me like there are no predictions of Br or bentazone concentrations for this initial period, presumably because no drainflow was predicted. The authors should be more explicit about this here.

Yes, that is what we meant to say. There is no drainflow simulated in the first period, which of course leads to the fact that no concentrations are predicted. This is most probably due to

the lack of soil moisture data below 1m depth, which led to erroneous initial moisture conditions, especially the initial depth of the water table. We will make this clearer in the revised version.

Technical Corrections

P10464 Are GCMs called Global climate models these days? They used to be known as General Circulation Models.

Both names are used nowadays within climate modelling. Originally, GCM was the abbreviation for 'general circulation model', but as these models are in recent decades often used for simulating global climate, GCMs have frequently been called 'global climate models' (e.g. Neelin 2011; Kjellström et al. 2011; World Meteorological Organization (WMO): http://www.wmo.int/pages/themes/climate/climate_models.php).

P10466 L 10 systemS

This will be changed in a revised manuscript.

Delta change method or delta method?

It seems that different names exist, Graham et al. 2007 wrote for instance: "The most common transfer method used to date has been the delta approach (e.g. [...]), often referred to as 'delta change' ". In other papers, the method is also referred to as change factor approach (Anandhi et al 2011).

Re GCMs. At some point references should be given for the GCMs used in the RCM scenarios presented in Table 2. Have I missed something?

No, we did not give any references for the GCMs, since we only got the RCM-outputs from the Swedish Meteorological and Hydrological Institute (SMHI) and just cited the reference where they used this ensemble. However, we can add appropriate references in Table 2, according to those given in Kjellström et al. (2011).

P10469 Why was relative humidity kept unchanged? Might help the nonmeteorologically minded reader, like myself!

Both observations and model results show that relative humidity is broadly maintained in a changing climate as changes in water vapour are governed by changes in temperature following the Clausius-Clapeyron relation (Bengtsson 2010). We will comment on this in a revised version of the manuscript.

P10469 L 28 "rather" not needed.

OK, this will be changed in a revised version.

P10470 Nice to see application date included.

Yes, since the choice of application dates is especially critical for pesticide losses in macroporous soils, we wanted to ensure that we did not add additional uncertainty due to the specific (random) choice of application dates.

P10478 L 5 rather – better approximately.
L17 strengthened
L16-19 sentence not clear. Consider rewriting.

We will include your suggestions and rewrite the sentence (L16-19) as we agree that it was not so clear.

Anonymous Referee #2
Received and published: 31 October 2013

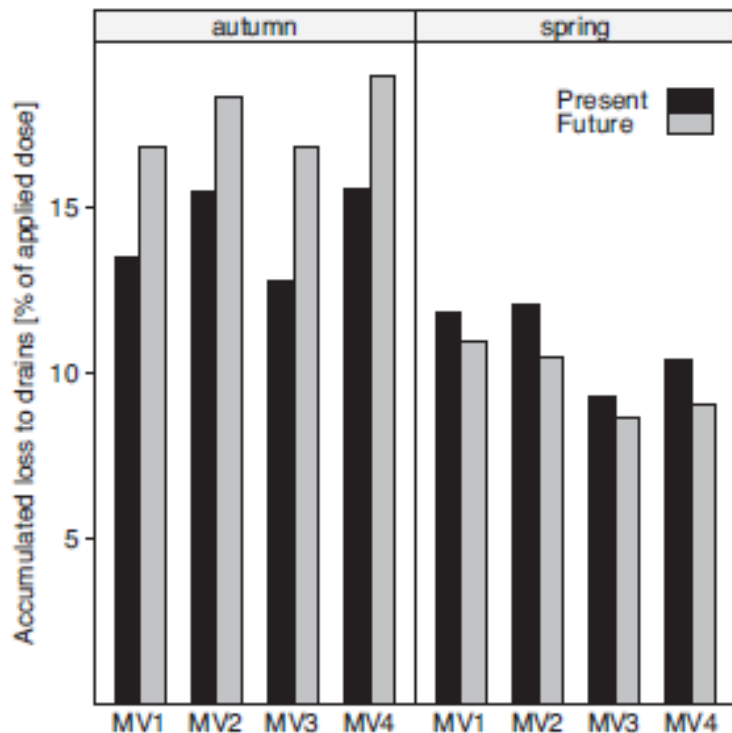
This is a well written paper about a relevant topic: Pesticide leaching under future climate scenarios. The major objective of the paper was elucidate uncertainties arising from different climate scenarios versus parameterization uncertainties. The study supports earlier findings that parameter uncertainty is less important than climate uncertainty.

The manuscript may be published almost as it is. I have just a few remarks the authors may want to consider in minor revisions.

- While the authors are correct with respect to decreasing sorption and increasing diffusion rates, this effects are probably very minor if temperature changes are just by a few degrees – here a quantitative statement about expected changes would be appreciated. In general, the authors could work out more clearly the physical reasons for the observed increased or reduced losses of pesticides applied in different seasons.

In a previous study (Steffens et al. 2013), we compared model structural uncertainty with parameter uncertainty and showed the magnitude of changes in pesticide leaching after spring and autumn applications, respectively, for different models that either included temperature dependent diffusion, or temperature dependent sorption, or both or neither (see the graph below, where we plotted the median losses for a weakly sorbed pesticide (bentazone) simulated with the different model versions (MV1-MV4), where MV1 is without temperature dependent functions, MV2 includes temperature dependent diffusion, MV3 temperature dependent sorption and MV4 both temperature dependent sorption and diffusion. MV4 is the model we used in the current paper). The effect of temperature dependent sorption seems to have a larger impact on pesticide leaching than diffusion. Especially for autumn application, the increase of temperature by a few degrees results in increases of 3-5% loss to drains as % of applied dose, which is not negligible.

The physical reasons for the contrasting effects depending on the application season are rather complex, but changes in losses after spring application are dominated by an increase in temperature during summer, when degradation takes place, whereas losses after autumn application are dominated by changes in autumn/winter precipitation.



- Change in soil moisture such as extended droughts would have much more significant effects concerning reduced (or stopped) biodegradation and faster gas diffusion in strongly unsaturated soil (leading to potentially strong volatilization of pesticide to the atmosphere). Also, during very dry periods strong sorption onto dry mineral surfaces may become significant besides partitioning into soil organic matter. All that is probably not considered by the model. Mentioning physical processes not covered by the model could clarify that for the readers.

Yes, these processes can affect the fate of pesticides. The direct effect of drought on degradation is considered in our model, as the degradation rate depends on temperature and soil moisture. The other processes mentioned by the reviewer are not considered in the model so an analysis of their effects is beyond the scope of the paper. Extreme droughts are not likely to be an important issue in Sweden, not even in the future. Nevertheless, as suggested by the reviewer, we will briefly mention in the introduction a few more of these processes that can influence pesticide fate and transport in soils to give a more complete picture.

References

Anandhi, A., Frej, A., Pierson, D.C., Schneiderman, E.M., Zion, M.S., Lounsbury, D., and Matonse, A.H.: Examination of change factor methodologies for climate change impact assessment. *Water Resour. Res.*, 47, W03501, doi: 10.1029/2010WR009104, 2011.

Bengtsson, L.: The global atmospheric water cycle. *Environ. Res. Lett.*, 5, doi:10.1088/1748-9326/5/2/025002. 2010

Beven, K. and Binley, A.: The future of distributed models: model calibration and uncertainty prediction, *Hydrol. Process.*, 6, 279–298, doi:10.1002/hyp.3360060305, 1992.

Boesten, J. J. T. I.: Sensitivity analysis of a mathematical model for pesticide leaching to groundwater. *Pestic. Sci.*, 31, 375–388, 1991.

Dubus, I.G., Brown, C.D.: Sensitivity and first-step uncertainty analyses for the preferential flow model MACRO. *J. Environ. Qual.*, 31, 227–240, 2002.

Dubus, I.G., Brown, C.D., and Beulke, S.: Sensitivity analyses for four pesticide leaching models. *Pest Manag. Sci.*, 59, 962–982, doi: 10.1002/ps.723, 2003.

Ghafoor, A., Jarvis, N.J., and Stenström, J.: Modelling pesticide sorption in the surface and subsurface soils of an agricultural catchment. *Pest Manag. Sci.*, 69, 919–929, doi: 10.1002/ps.3453, 2013.

Graham, L. P., Andréasson, J, and Carlsson, B.: Assessing climate change impacts on hydrology from an ensemble of regional climate models, model scales and linking methods – a case study on the Lule River basin, *Climatic Change*, 81, 293–307, doi:10.1007/s10584-006-9215-2, 2007.

Kjellström, E., Nikulin, G., Hansson, U., Strandberg, G., and Ullerstig, A.: 21st century changes in the European climate: uncertainties derived from an ensemble of regional climate model simulations. *Tellus*, 63, 24–40, doi: 10.1111/j.1600-0870.2010.00475.x, 2011.

Larsbo, M., and Jarvis, N.J.: Simulating solute transport in a structured field soil: Uncertainty in parameter identification and predictions. *J. Environ. Qual.*, 34, 621–634, doi:10.2136/vzj2004.0137, 2005.

Larsbo, M., Roulier, S., Stenemo, F., Kasteel, R., and Jarvis, N.J.: An improved dual-permeability model of water flow and solute transport in the vadose zone. *Vadose Zone J.*, 4, 398–406, 2005.

Liu, Y., Freer, J., Beven, K., and Matgen, P.: Towards a limits of acceptability approach to the calibration of hydrological models: Extending observation error. *J. Hydrol.*, 367, 93–103, doi:10.1016/j.jhydrol.2009.01.016, 2009.

Neelin, J. D.: *Climate change and climate modelling*. Cambridge University Press, Cambridge, UK., 282pp, 2011.

Nylinder, J., Stenberg, M., Jansson, P.E., Klemedtsson, A.K., Weslien, P., and Klemedtsson, L.: Modelling uncertainty for nitrate leaching and nitrous oxide emissions based on a Swedish field experiment with organic crop rotation. *Agric. Ecosyst. Environ.*, 141, 167–183, doi:10.1016/j.agee.2011.02.027, 2011.

Steffens, K., Larsbo, M., Moeys, J., Jarvis, N., and Lewan, E.: Predicting pesticide leaching under climate change: Importance of model structure and parameter uncertainty. *Agric. Ecosyst. Environ.*, 172, 24–34, doi: 10.1016/j.agee.2013.03.018, 2013.

World Meteorological Organization (WMO):
http://www.wmo.int/pages/themes/climate/climate_models.php