

Interactive comment on “Modeling subsurface transport in extensive glaciofluvial and littoral sediments to remediate a municipal drinking water aquifer” by M. Bergvall et al.

M. Bergvall et al.

martin.bergvall@tyrens.se

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We thank Dr. habil. Köhne for very helpful, constructive and insight full comments which will substantially improve the readability of the paper.

Comment: After reading the paper I [Dr. habil. Köhne] asked myself, what can we really learn from this study, for instance, about processes and their modelling at this scale? Partly this question may be caused by missing information. I will focus on the vadose zone modelling approach, since I found this less clear than the groundwater part.

Reply: The modeling results show that in coarse-grained settings it is not necessary to model all possible processes in the vadose zone in order to predict concentrations in groundwater at a regional scale. For example, possible forms of preferential flow can be modeled by large macrodispersion, and it is probable that local variability in mass fluxes within a contaminated area is mixed at regional scale. To better explain our results we would add Figs. A, B, C and D (see below), and clarify the methods and discussion sections. For example we would revise the discussion section and introduce it with the following: ‘The results were received from a glaciofluvial esker aquifer contaminated by the herbicide degradation product BAM. Despite the herbicide has been modestly used, BAM has contaminated an aquifer holding large groundwater quantities, and particularly two extraction wells located about 1.5 km down-gradient from the source have concentrations that exceed the European drinking water standard for pesticides (0.1 $\mu\text{g l}^{-1}$). At regional scale, the model was able to explain observed BAM concentrations in the groundwater, and to evaluate the effect of remediation strategies.’ Further improvements are found below in our replies to the comments.

1. (1) The vadose zone model was said to be assumed to represent the ‘total open land area’. Figure 1 also gives the impression there was only one 1d-model for the entire domain. In reality there was pesticide input from the nursery and none from outside areas. If you apply a mixture (4% of 0.17 g m⁻²) for the total area instead of applying it only at the tree nursery area, this is a drastic deviation from the real situation! But looking at Fig. 6, simulated BAM does originate from the nursery. Please clarify in the text!

Reply: To avoid confusion with the expression ‘total open land area’ we would revise the text to ‘total nursery area’. The vadose zone model was only applied to the nursery field, which we would clarify in Fig. 1 and in the manuscript. We would also insert a reference to Fig. 1 at P1738 L10.

2. (2) Data of 5 measured soil profiles were mixed to give a representative soil profile. However, does that really represent average soil properties for the ‘total land area’

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(see above)? How can this approach be justified? I think the calibration adjusted the parameter of this profile, but this then will no longer represent any measured data, but an ‘effective profile’.

Reply: First of all ‘total open land area’ refers to the open land area of the nursery (see above), and not to the vadose zone above the whole groundwater zone. The nursery area is 12 ha and within this area we found similar layering of sandy sediments (according to six soil profiles in total). The model parameters of the ten soils found in Table 3 are based on analyzed soil textures in the 10 m deep soil profile, but calibrated to match measured water content at various depths. The water content was measured every 20 cm in the 10 m deep soil profile by the oven-drying method. The water content was also measured on thirteen occasions with a depth moisture probe. The water content data obtained by the oven-drying method were used to calibrate the model, and the depth moisture probe data was used to validate the model. We would clarify this in sections 2.3 and 2.5.1. We would also add Fig. A, which shows observed and simulated water content in the soil profile.

3. (3) The parameterization of the vadose zone: Was K_s measured? Indicate in Table 1 (e.g., using superscript symbols) which quantities were measured, which calibrated, and which represent literature estimates. Add dispersivities to Table 1.

Reply: From the analyzed soil textures K_s was calculated by the Hazen method. However, the values found in Table 3 are calibrated values. We assume that you refer to Table 3 and not Table 1, and would revise Table 3 according to your suggestions. However, in Table 1 we would add the Hazen calculated K_s values.

4. (4) The role of the vadose zone modelling remains almost in the dark. The calibration gives some answers, but results for simulation and measurement should be shown also in terms of water contents and pesticide concentrations in the vadose zone.

Reply: When we chose figures for the previous version of the manuscript we focused on the extraction wells. However, we realize now that the vadose zone model needs

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to be better explained. We would add Fig. A with observed and simulated water contents as shown above. At P1739 L27 we would add the following: ‘The root-mean of squared differences for the validation data gave a corresponding error of 24 %. The infiltration rate and the generated bottom water flux at 10.2 m depth are shown in Fig. B (see below). The figure shows a seasonal variation of the bottom flux.’ We would add Figures B, C and D that show infiltration rate, observed and modeled dichlobenil concentrations, and simulated water flux, mass fluxes and concentrations of BAM at the bottom of the model. The analyzed dichlobenil concentrations found in Fig. C would also be added to Table 1.

5. (5) The beginning of the discussion of the results it is stated that a simpler approach leads to the same transit times for water and BAM. As it is now, the discussion is of limited value since the reader does not know the study by Bergvall et al. (2007). Either this part of discussion should be deleted, or more information on the ‘mass-balance model’ (use different term? Note that Richards and CDE are also mass-balance equations) used in Bergvall et al. would be needed.

Reply: The model by Bergvall et al. (2007) is better referred to as a stationary, non-distributed model. At P1745 L11, we would add more information about the model: ‘To model transport in the vadose zone, Bergvall et al. (2007) applied a simplified chromatographic flow model, implying that the infiltration rate and the total amount of water were constant. A retardation factor was used to account for sorption. To model transport in groundwater, Bergvall et al. (2007) used Darcy’s law at five cross-sections along the flow direction between the nursery field and the two down-gradient extraction wells (Fig. 2). Dispersion was not directly modeled, but indirectly represented by the use of Monte-Carlo simulations to calculate contaminant transport within a range of values to give various outcomes. The degradation rate of dichlobenil was not represented in the model by Bergvall et al. (2007), and the results were highly dependent on sorption related coefficients.’

6. (6) Could you show simulated time series of concentrations and fluxes at the 10-m

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deep bottom of the vadose zone, serving as input to the GW model? Perhaps these can be compared to simulation results of the mass-balance model. Perhaps mean transit times are similar, but peak events differ. How could this affect overall transit times towards the extraction wells? Can you better demonstrate the benefit of using your coupled approach?

Reply: We would add a figure showing concentrations and fluxes at the bottom of the vadose zone (see above, Fig. D). At the bottom of the vadose zone the mass fluxes of BAM (Fig. D) corresponded with the variation of water fluxes (Fig. B), and resulted in stable BAM concentrations with small variations from day to day (Fig. D). With the groundwater model we also studied the transport of a solute pulse with ten times higher concentration compared to the mean. A 1-day long pulse was applied in August 2000 (a year with many heavy rainfalls) as input to the groundwater model. The pulse became clearly visible in the extraction wells and had in 2004 a maximum concentration, which was $0.20 \mu\text{g/l}$ higher compared to the presented model result. When we applied a similar pulse, but 30-day long, it resulted in, at the most, $0.23 \mu\text{g/l}$ higher concentration in the extraction wells compared to the presented model result. Comparing these results to the variability of the analyzed BAM concentrations we believe that local variability in mass fluxes will be mixed at regional scale. At P1744 L25 the following text would be added: 'The transit time of groundwater from the northern to the southern end of the nursery was calculated to be more than one year, implying that local variability in mass fluxes will be mixed at regional scale.' We would better explain the benefit of using our coupled approach, and the following text would be added at P1745 L20: 'Despite the prediction ability of the simple mass-balance model in coarse-grained settings, the distributed numerical model should be used if the contaminant transport needs to be calculated more accurately, e.g. to predict concentrations at a specific point or to evaluate remediation strategies.'

7. (7) Although using daily time steps may be a high time resolution for this long-term study, in terms of process time scale it is not. If daily averages are used, the rainfall

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rate will less often exceed K_{sat} (assuming measured value) of the soil than if real rates (e.g., hourly time steps) would be applied. Thus, saturation of soil layers is not reached or less often. The higher sensitivity to infiltration than to $K_{sat}(\text{soil})$, reflects this. So overall the ranking of sensitivities is meaningful only within the (time scale of) this modelling approach.

Reply: At P1739 L22 the following text would be added: ‘During the simulation time, the maximum calculated infiltration rate was 40 mm day⁻¹, and occurred a day when the precipitation was 42 mm day⁻¹. Since this is 103 times smaller compared to the saturated hydraulic conductivity at the soil surface (Table 3), and rain with an intensity of 20 mm per half an hour occurs only once in ten years at the field site (SMHI, 2009), daily time resolution of input data was assumed to be satisfactory. Consequently, we considered matrix flow to be the dominant process.’ We would also revise the sentence at P1743 L13 to: ‘Considering the model was run at daily time resolution, the saturated hydraulic conductivity of the aquifer was found to be the most sensitive parameter for determining the predicted concentration of BAM in the extraction wells (Table 4).’ At P1743 L17 we would add: ‘Since the vadose zone model was applied on a coarse-grained unstructured soil with high saturated hydraulic conductivities exceeding normally occurring infiltration rates by orders of magnitude, the validity of the results from the sensitivity analysis might be extended to time scales smaller than one day.’

8. It seems that macropore flow effects were not required to explain the observations. Although calibrated large macrodispersivity values in the unsaturated zone point to non-ideal transport. The macropore flow discussion could be shortened (or skipped altogether).

Reply: We agree and would skip the discussion of macropore flow. Instead we would add the following text to the discussion section: ‘On the groundwater table underlying the source the breakthrough curve of BAM became wide (Fig. 4). Similar results have been obtained in previous studies on pesticide leaching in unstructured soils (Kjaer et al., 2005; Brown et al., 2000). Since sandy soils naturally contain a high num-

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ber of large pores, they permit infiltration when exposed to heavy rainfall. A lack of macropores implies that matrix flow is the dominant process, which causes a wide breakthrough curve. This helps to explain why the BAM contaminant was predicted at levels exceeding the drinking water standard at the two extraction wells for three to four decades.’ The text at P1746 L23-27 would be moved to the results section at P1743 L18-23. In the discussion the text at P1746 L23-27 would be replaced by the following text: ‘The observed and simulated concentrations were not perfectly matched. Reasons can be both that all significant transport processes were not fully understood and included in the model, and that the quality of the analyzed BAM samples in groundwater varies. In particular, the sampling quality may vary in those samples that were collected earlier than 2007. Since the model was run at regional scale combined with a deep vadose zone and coarse texture, it was assumed that preferential flow was insignificant. For example, it is not evident whether fingered flow can be a significant process at infiltration rates that normally occur at this site (Selker et al., 1992). However, any possible forms of preferential flow in the vadose zone were incorporated in the model through the dispersion term, using apparently higher macrodispersivity values than the true dispersion.’

9. The moving average procedure in Fig. 4 is unclear.

Reply: We would explain the moving average procedure by adding the following text at P1737 L3: ‘Fig. 4 also shows a moving average, which is the mean value of the closest four observations, before and after each observation. This means that the integral of the observed concentrations should over time equal the integral of the moving average concentrations.’

10. The contrasting concentration patterns in Fig. 5 for west and east wells could be explained better, e.g., by using graphical representations of the plume and velocity vectors in and between lumping periods.

Reply: We agree that it would be interesting to include a figure showing the contami-

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nant plume and velocity vectors when the extraction wells are closed, and when they are running. However, in order to improve the explanation of the vadose zone model we have increased the number of figures by four. We would therefore prefer to not include additional figures. However, we would add the following text at P1747 L9: ‘According to the model the explanation is that the capture zone of the east well is partly located in finer and less permeable material (Fig. 3).’

Comment: Extra need for clarification is also suggested below in the specific comments. I [Dr. habil. Köhne] suggest that this study can be published in HESS after revision.

SPECIFIC COMMENTS 11. ABSTRACT Line 4: In formerly glaciated areas – regions

Reply: We agree

12. Line 7: subsurface transport of pesticides in extensive glaciofluvial and littoral sediments – not soil?

Reply: The model was applied to an unstructured soil, with no developed horizons and with low organic carbon content. At least the model results are applicable to glaciofluvial and littoral sediments, but probably the applicability can be widened to glaciofluvial and littoral soils. At P1735 L25 the following text would be added: ‘The soil has not been developed but mechanically cultivated, and the organic carbon content is low.’

13. Line 9: point scale – or soil profile scale.

Reply: Soil profile scale. We would change this throughout the manuscript.

14. Page 1731, Line 4: As part of a national monitoring program, 28% of the groundwater supplies investigated between 2005 and 2008 were [add: identified as being] contaminated: : :

Reply: Thank you

15. Page 1731, Line 11: “approximately one hundred groundwater supplies were re-

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cently closed due to BAM contamination” - what means “supplies” – extraction wells?
Certainly not entire waterworks?

Reply: Thank you. We would change to ‘groundwater extraction wells’.

16. Page 1731, Page Line 15: “Dichlobenil has largely been used as a pesticide” – was mostly used as a herbicide

Reply: We agree

17. Page 1732, Line 15: “The lack of models for eskers : : :” define eskers, since this may not be known to all readers. I wonder if this statement is accurate: not most aquifers will be eskers even in regions that were glaciated during the last ice age – there are other types of glaciofluvial sediments that are widespread? So make statement more general?

Reply: At P1732 L12 we would add: ‘An esker is a narrow ridge of gravelly and sandy glacial material originally deposited by a stream in an ice-walled valley.’ The model approach will apply to similar settings. At L15 we would make the statement more general and replace ‘esker’ by ‘glaciofluvial sediments’.

18. Page 1733, Line 6: “: : :the calculated pesticide concentration from the vadose zone model was used as the input” – questions on the coupling: you would rather need both concentration in percolate and volume of water exchanged; so the mass flux. Please clarify. Other questions: coupling at what depth? Time resolution of one day? Was water flow also used as input to Modflow/MT3DMS via sink/source term? Maybe add ‘see below’ or the like, if these things are explained in a later section.

Reply: We would prefer to present the general modeling approach in section 2.1 and then present the details of the application in section 2.5. In our response to comment no. 7 we address the time resolution with new text added at P1739 L22. In order to clarify the manuscript we would prefer to remove the sentence that addresses time resolution in section 2.1 at P1733 L18-20. At P1733 L6 we would revise the text to:

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‘...the calculated pesticide concentration from the bottom of the vadose zone model was used as the input, through the recharge parameter, to the 3D finite-difference model MODFLOW (Harbaugh et al., 2000) coupled with MT3DMS. In our response to comment no. 4 we have added Figs. B and D. At P1739 L15 we would add the following text: ‘The vadose zone model was calibrated with data sampled at the identified hotspot (Fig. 2). Considering the higher dichlobenil concentrations found at the hotspot, the application rate was assumed to be 1.7 g m⁻² in the calibration model.’ At P1739 L27 we would revise the sentence to: ‘The solute component of the model was calibrated to achieve the best match between the calculated and observed hotspot-concentrations of dichlobenil in the solid and solute phases (Table 1, Fig. C)’. At P1740 L2 we would add the following text: ‘Concentrations and mass fluxes of BAM that were generated by the prediction model at the bottom of the vadose zone are shown in Fig. D. At the bottom of the vadose zone the mass fluxes of BAM corresponded with the variation of water fluxes (Fig. B), and resulted in stable BAM concentrations (Fig. D). In comparison with the mass fluxes of BAM from the calibration model, the prediction model generated correspondingly lower mass fluxes equivalent to the lower application rate, i.e. in the predictions it was valid to use the calibration model with a lower application rate.’

19. Page 1734, Line 8: “: :theta is the porosity” – in eqs. 1 and 2 theta denotes water content. Use different symbols?

Reply: Thanks. We would revise the theta symbol in Eq. (4) to θ_k , where θ_k is the kinematic porosity used for solute transport in groundwater.

20. Page 1734, Line 13: “To evaluate the robustness of the model’s predictions” – what means “the model” – eqs. (1) through (4) (Hydrus+Modflow+MT3DMS)?

Reply: We would add a reference to Eqs. (1) up to (4).

21. Page 1734, Line 13: “were perturbed with changes of ± 2 to 10%, corresponding to the observed variability (Bergvall et al., 2007)” – is not this quite a small variability, thinking of, for example, infiltration into soil and hydraulic conductivity of soil or aquifer?

Natural rainfall intensity and such parameters can vary much more. Please explain.

Reply: We would remove the reference and revise this sentence to: ‘To calculate the effect that a parameter has on the model outcome, the parameters in the calibrated models were perturbed with changes of ± 2 to 10 %.’

22. Page 1735, Line 24: “The former nursery is situated on well sorted, unstructured, littoral sand and lenses” – what about the soil type and physical/chemical soil properties at this site? Soil properties have a strong effect of sorption and degradation.

Reply: In our response to comment no. 12 we partly addressed this topic. The soil is a cultivated spodosol with low content of total organic carbon (1.0 to 6.3 g kg⁻¹). In aerobic sandy sediments with TOC less than 1 g kg⁻¹ iron oxides or other minerals contribute to the sorption of both dichlobenil and BAM (Clausen, 2004). Sorption has been found to correlate strongly to TOC for both dichlobenil and BAM ($R^2=0.91$), especially when TOC is higher than 1 g kg⁻¹ (Clausen, 2004). In sandy sediments when TOC is less than 1 g kg⁻¹ other factors may contribute to the sorption, such as iron oxides and clay minerals (Clausen, 2004). We had almost no clay minerals present and our TOC was not less than 1 g kg⁻¹, why we assumed that sorption was correlated to TOC. The application of the modeling approach, including sorption and degradation, is explained in detail in section 2.5.1.

23. Page 1736, Line 9: “At one identified hotspot (Fig. 2)” – I could not locate this hotspot in Fig. 2, apply symbol and indicate in legend?.

Reply: We see that the hotspot-text is hidden among the exposed bedrocks in the northeastern part of the map. To improve the visibility we would put the hotspot-text outside the exposed bedrocks.

24. Page 1737, Line 13: “The mean leakage was estimated to be 5% of the mean discharge rate of 160 l s⁻¹ (SMHI, 2009).” Does this 5% loss significantly exceed the error margin of the discharge rate measurement?

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Reply: Using multiple measurements the precision was always better than 5% using the salt dilution technique. The technique was used at four occasions to measure the stream discharge. At each occasions three measurements were conducted. In total, all twelve measurements showed that there is a leakage from the stream.

25. Page 1738, line 3: “Assuming a Gaussian distribution, the longitudinal dispersion ranged between 0.2m and 1.5 m, at the 2m scale.” Gaussian distribution (in horizontal direction!?): what assumption is underlying here – advection dispersion? But is 1.5 m dispersivity within 2 m transport distance not rather long for AD behaviour?

Reply: We would revise this sentence to: ‘Assuming Gaussian distribution in the flow direction, the longitudinal dispersion was calculated by an analytical solution of the Fickian advection-dispersion model of solute transport, and the resulting values ranged between 0.2 and 1.5 m, at the 2 m scale.’ We agree that a dispersivity value of 1.5 m at the 2 m scale is rather high. However, the calibrated dispersivity value at the 20 m grid-cell scale was 5 m, which also is rather high. At P1742 L17 we used Eq. (6) to calculate dispersivity values at the different scales. The empirical equation suggests similarly lower dispersivity values at both scales, which we find consistent. Nevertheless, in comparison with advection the effect of dispersion is low. Before the sentence at P1746 L28 in the discussion section we would add the following: ‘In groundwater the results showed a limited lateral dispersal of the contaminant BAM, as it moves from the source in littoral sand to the glaciofluvial esker, which has a comparatively coarser texture and higher hydraulic conductivity.’ In comparison to a Finnish study with similar settings, our longitudinal dispersivity values are low. In the discussion section at P1747 L4 we would add the following: ‘The limited lateral dispersal in the esker probably explains why the highly water soluble degradation product BAM can migrate in high concentrations over considerable distances. However, in comparison with an esker aquifer modeled by Artimo (2002) in Finland at similar scale, the longitudinal dispersivity values were 8 to 10 times smaller. Artimo did, however, not include the vadose zone in the modeling, and the aquifer was contaminated by the dense non-aqueous phase

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liquid tetra-chloroethylene, which makes further comparisons difficult.'

26. Page 1738 – Line 6: “The decoupling of the vadose and groundwater zones was found to be appropriate since there was little variation in the groundwater table depth” – Decoupling - does this mean no GW recharge through the vadose zone was assumed? How does then the solute get into the GW model, is it just added as mass?

Reply: Decoupling was not a good word to use. We would replace the sentence at P1738 L6-8 by the following: ‘The vadose zone model was only applied to the nursery area (Fig. 2). Since there was little variation in groundwater level within the nursery area (standard deviation = 0.06 m, Table 1), we assumed no feedback from the groundwater model to the vadose zone model.’

27. What (is the range in) surface area(s) that was (were) represented by the point-scale model? Was every grid cell of the subsurface model coupled with the 1 D model, or were the hydrologic units? Explicitly mention narrow grid spacing of 2 cm (which is sufficient resolution for valid calculation: Vogel, H.-J., Ippisch, O., 2008. Estimation of a Critical Spatial Discretization Limit for Solving Richards’ Equation at Large Scales. Vadose Zone J., 7(1): 112-114).

Reply: Thanks for a nice reference. We would include it in the manuscript at P1738 L16: ‘The grid was narrowly spaced at 2 cm in the vertical direction, which is sufficient resolution for valid calculations of sandy soils (Vogel & Ippisch, 2008).’ See also our responses to comments no. 1 and 26.

28. Page 1738 – Line 18: “Ten soils with various properties were defined to describe the vadose zone, ranging from fine sandy silt to coarse sandy medium sand (Table 3).” Add: “The information underlying Table 3 was obtained as follows”. Some measurements were already mentioned above, but it is not clear what information entered the table – including parameters for chemical reactions. The remainder of this page reads like a discussion of the validity of the data, but does not explain how they were derived, which I found confusing. Adding some explicit statements in this part would be helpful.

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Reply: We would add superscript letters to show which parameters that were measured and calibrated. We would revise the sentence at P1738 L20 to: ‘Degradation of dichlobenil and BAM was described by first order kinetics. Conditioned on the total mass balance, the half-lives were calibrated to the values given in Table 3.’ See also our responses to comments no. 29 and 30.

29. Page 1739, Line 1: “As moderate concentrations of dichlobenil were found at depths greater than 0.9 m, we assumed that the linear Freundlich sorption isotherm was applicable.” To make immediately clear that linear isotherm was used only for the depth below 0.9 m, I suggest to restate “At depths greater than 0.9 m, moderate concentrations of dichlobenil were found, so we assumed that the linear sorption isotherm was applicable.” However, I do not understand the proposed relation. Why should moderate concentrations point to a linear isotherm?

Reply: We would revise the sentences at P1739 L1-5 to: ‘It was assumed that the Langmuir sorption isotherm was applicable. However, as the linear Freundlich isotherm produced similar results at depths greater than 0.9 m, the Langmuir isotherm was not used at these depths in order to save computing time. Clausen et al. (2004) found a linear correlation between the total organic carbon content (TOC) and sorption of dichlobenil and BAM as long as the TOC > 1.0 g kg⁻¹: (7) (8) where K_d_{diklo} and K_d_{BAM} are the sorption coefficients of dichlobenil and BAM, respectively; and f_{oc} is the weight fraction of TOC. The sorption coefficients for dichlobenil and BAM were initially calculated in accordance with Eqs. (7) and (8), but were later calibrated to improve the correlation with measured concentrations of dichlobenil in the solid and dissolved phases (Table 3). About 20 cm of the topsoil had been excavated before we sampled the soil, and since there had been vegetation we assumed about 3 to 5 times higher TOC-content in the topsoil layer (Table 3) compared to our analyzed maximum TOC (Table 1).’

30. Page 1739, Line 5: “Dispersivity was initially assumed to be 10% of the soil-profile depth, but was calibrated to 5 m, except for the top meter which was calibrated to 1 m.”

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Can you squeeze in dispersivity values (an important model parameter) into Table 3? Indicate a possible physical interpretation of these extremely large dispersivity values of up to 5 m?

Reply: We would add the dispersivity values to Table 3. We would also revise the sentence at P1739 L5-7 to: ‘We assumed high macrodispersivity values to account for possible forms of preferential flow. Dispersivity, α_{Lq} , was initially assumed to be 10 % of the soil-profile depth, but was calibrated to 5 m except for the top meter.’

31. Page 1739, Line 23: “The van Genuchten-Mualem parameters were calibrated against water content measured at 51 depths on 13 occasions. The mean error of the total water balance was 1.3%.” If the water balance was known, why was this information not used in the model calibration to better constrain the model? Was the calibration for water and solute done sequentially (literature says its better to do it simultaneously to utilize mutual dependencies). And the statement does not tell if this calibration is for one point only, or for several sites – the profile hydraulic properties certainly show spatial variability?

Reply: We would add Fig. A to clarify how the flow model was calibrated. We would revise the text at P1739 L23-27 to: ‘The water flow and the solute transport models were calibrated sequentially. The van Genuchten-Mualem parameters including saturated hydraulic conductivity were calibrated against water contents analyzed by the oven-drying method, and then validated with the moisture probe data obtained at the hotspot (Fig. A). The mean error of the total water balance was 1.3 %. The root-mean of the squared differences between the calculated and observed water contents obtained by the oven-drying method was normalized by the observed range, giving a calibration error of 10 %. The root-mean of squared differences for the validation data gave a corresponding error of 24 %. The infiltration rate and the generated bottom water flux at 10.2 m depth are shown in Fig. B. The figure shows a seasonal variation of the bottom flux.’

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32. Page 1739, Line 27: “The solute component of the model was calibrated to achieve the best match between the calculated and observed concentrations of dichlobenil and BAM, in the solid and solute phases.” Fitting which parameters? Fitted at what place – an average of the 5 sampling locations at nursery area? If stated above, briefly repeat here for clarity. Perhaps refer to results from sensitivity analysis (some ‘see below’ statement) used to identify the important parameters to be fitted.

Reply: We would add Fig. C to clarify how the solute model was calibrated. We would revise the text at P1739 L27-P1740 L2 to: ‘The solute component of the model was calibrated to achieve the best match between the calculated and observed hotspot-concentrations of dichlobenil in the solid and solute phases (Table 1, Fig. C). The sensitivity analysis that was undertaken, Eq. (5), helped to identify the important parameters to be fitted. Since the observed concentrations had high variability the obtained fit to simulated concentrations was only in the same order of magnitude. The root-mean-squares of the residuals of the adsorbed and liquid phases were normalized by the observed concentration range giving calibration errors of 38 % and 39 %, respectively. Concentrations and mass fluxes of BAM that were generated by the prediction model at the bottom of the vadose zone are shown in Fig. D.’

33. Page 1740, Line 8: “Initially, the groundwater table was calculated in a one-layer model with the same boundary conditions.” What was the result of using one layer and why was it discarded? If not important, delete statement.

Reply: We would delete this statement.

34. Page 1740, Line 20: “Four different recharge rates were defined for the model: most areas had a constant net infiltration rate of 8.2×10^{-4} m d⁻¹.” Does this number correspond to what is simulated with the 1 D model at 10.2 m depth? If there is a considerable difference, this affects the simulated loss of pesticide to the GW.

Reply: We would replace the sentence at P1740 L21-23 by the following: ‘The constant rate at the nursery field was justified by the 10 m thick vadose zone, which smoothes

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out variations in the net infiltration rate and changes in the groundwater level (Fig. B, Table 1). The BAM concentrations generated by the vadose zone model had also small variations from day to day (Fig. D).

35. Page 1742, Line 1-3. “The simulation started on 1 January 1976 and the calibration model was run for 33.5 yr until 1 July 2009. The prediction model was run for 45 yr until 31 December 2020.” – Was the vadose zone model also run for this time and if not, how was the coupling for times covering different simulation periods done?

Reply: We would clarify this in the description of the vadose zone model by adding the following at P1739 L16: ‘The simulation started on 1st September 1975 and the calibration model was run until 1st November 2003. The prediction model was run for 45 years until 31st December 2020.’

36. Page 1742, Line 1-3. “Calibration of dispersivity in the three dimensions was conducted manually” – why, was there no automated calibration for solute transport available?

Reply: When calibration of the solute transport module was conducted we had no automated calibration tool available.

37. Page 1742, Line 18: “Equation (6) gives a longitudinal dispersivity of 0.96m at the grid cell scale” if that is a correct application of Eq. 6 to use it for the grid cell size of 20 m (and not for the entire aquifer dimension), then what is it supposed to describe – numerical or physical dispersion?

Reply: We applied Eq. (6) to compare physical dispersion at different scales. Both the measured and the calibrated dispersivity values turned out to be higher than expected from Eq. 6. We have also addressed this topic in our response to comment no. 25. We would revise the sentence at P1742 L13-16 with the following: ‘This is consistent with the results from the local-scale, salt-injection study (α_L between 0.2 and 1.5 m), and can be put into perspective with the empirical relationship between longitudinal

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dispersivity (αL) and field scale (L), suggested by Schulze-Makuch (2005)'. We would replace the sentence at P1742 L18-19 by the following: 'Equation (6) gives a longitudinal dispersivity of 32 m at the field scale ($L=1500$ m), 0.96 m at the grid-cell scale ($L=20$ m), and 0.15 m at the scale of the salt-injection study ($L=2$ m). Both the calibrated and the measured longitudinal dispersivity values were comparatively higher than the values obtained from Eq. (6).'

38. Page 1742, Line 21: “: :and sorption is limited.” How does sorption reduce lateral dispersion (add reference?)

Reply: This part of the sentence would be deleted.

39. 1743, Line 13: “The saturated hydraulic conductivity “ add: “of the aquifer”

Reply: Yes

40. Page 1743, Line 15: “Together with the infiltration rate” D'accord, but one should be aware that the sensitivity of infiltration rate and van Genuchten parameters certainly depend on the time resolution of the model simulation. Although using daily time steps may seem as a high time resolution for this long-term study, in terms of process time scale it is not. If daily averages are used, the rainfall rate will less often exceed the K_{sat} of the soil than if real rates (hourly time steps) would be applied. The higher sensitivity to infiltration, than to $K_{sat}(\text{soil})$, may reflect this. So overall the ranking of sensitivities is meaningful only within the (time scale of) this modelling approach.

Reply: We have addressed this in our response to comment no. 7.

41. Page 1743, Line 18: “The simulated and observed concentrations, at one observation well at the nursery and at the extraction wells, are shown in Figs. 4 and 5.” – Please briefly explain moving average procedure in Figure 4

Reply: We have addressed this in our response to comment no. 9.

42. Page 1744, Line 13: “At the end of the predictive simulation,” – in 2012?

Reply: We would add 'in 2020'.

43. Page 1744, Line 20: "The simulation of the vadose zone gave a median transit time" – what sample does the median refer to – averaged over number of years? Or the number of 1D models linked to the GW model – but there seems to be only a single model, i.e. uniform conditions from upper boundary (was there no spatial variability in infiltration in the contaminated area)? Was the vadose zone model(s) only used in the contaminated area (tree nursery), while outside of this area, the upper boundary condition of MODFLOW was equal to prescribed GW recharge or recharge related to some leakage and conductance? I do not recall this being explained somewhere. At least this info could be added in Fig. 3.

Reply: We would clarify this by replacing the sentence at P1744 L20-21 by the following: 'To calculate transit times in the vadose zone model, the medians of the water content at each calculation node was used. The medians were received from daily output during one simulation year (1st July 2003 to 30th June 2004). At each calculation node the retardation factor and transit times of water and BAM could then be calculated, and resulted in a median transit time to groundwater of 4.3 years for water and 7.2 years for BAM.' At P1744 L23 we would add the following: 'In order to calculate transit times with the calibrated groundwater flow model, particle tracking was conducted from the groundwater table of the nursery field.'

44. Page 1745, Line 3: "and the simulation was run with pumping rates of 1.5 l s⁻¹ and 0.75 l s⁻¹." Why larger values in simulation than in test– explain.

Reply: We would revise the sentence at P1745 L1-3: 'During the test, the pumping rates in the two wells were 0.82 l s⁻¹ and 0.27 l s⁻¹. Since the evaluation of the pumping test indicated that more groundwater could be extracted, the simulation was run with pumping rates of 1.5 l s⁻¹ and 0.75 l s⁻¹.'

45. Page 1745, Line 17 "However, in the vadose zone the models gave similar median transit times for both water and BAM." – To avoid possible confusion in this statement

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(transit time of water similar to that of BAM), consider adding “with similar retardation factor of 1.7.” How is the transit time of the mass balance approach calculated?

Reply: We would add ‘with similar retardation factor of 1.7’. The calculation of transit times in the simple mass-balance model is addressed in our response to comment no. 5.

46. Page 1746, Line 1: “We considered whether macropore flow should be included in the model. During our simulation time of 33.5 yr, the maximum infiltration rate was 40 mm day⁻¹. Compared to the saturated hydraulic conductivity at the soil surface, this is 1000 times smaller and indicates that matrix flow is the dominant process.” Be careful, daily averages cannot be compared to values for K_s measured under ‘real-time conditions’. The main rainfall event on the day with 40 mm rainfall may have had an intensity of, e.g., 20 mm/h. OK, that is still much smaller than K_s at the surface, but in lower layers the situation is different.

Reply: We have addressed this in our responses to comments no. 7 and 8.

47. Page 1746, Line 5: “The pressure heads were studied with our model, but they were not higher than -0.75m at any time-step or depth.” – Again, that is caused by your choice of daily time steps, unless K_s values are somehow calibrated for exactly this time interval used in the boundary condition. Else if measured values are used, the conclusion is not correct. The next statement about coarse texture is a stronger argument against macropore flow, although other forms of preferential flow (fingering etc.) might still play a role.

Reply: We have addressed this in our response to comment no. 8.

48. Page 1746, Line 14: “apparently higher dispersion than the true dispersion.” Which you are already doing, right, using macrodispersivity values in the m range.

Reply: We have addressed this in our response to comment no. 8.

49. I wonder why you are discussing macropore flow in the first place? Were there

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any hints at macropore flow? There is no feasible way to test it experimentally for a 10 m deep vadose zone. While flow in the vadose zone is almost never one hundred percent chromatographic, I would assume from your description that this seems to be a relatively homogeneous flow case. This can indeed be represented by an increased macrodispersivity.

Reply: We have addressed this in our response to comment no. 8.

50. Page 1746, Line 24: “the integral of the observed concentrations” – integral over what giving what quantity; define this above, at first reference to Figure 4.

Reply: We have addressed this in our response to comment no. 9.

51. Page 1747, Line 6: “In this well, the concentration increases when the pumps are stopped because the contaminated water becomes less diluted (Fig. 5).” This will correspond to a shift in the flow direction to the natural one (after stopping pumping) favoring the west well, whereas the east well is partly (in Layer 2) outside the esker. Is that an explanation for the contrasting concentration patterns?

Reply: Yes, that is the explanation. We have addressed this in our response to comment no. 10.

52. Page 1747, Line 26: “be clean in four years.” Means: $c < 0.1 \text{ } \mu\text{g/L}$.

Reply: We would add the following at the end of the sentence: ‘(concentration $< 0.1 \text{ } \mu\text{g l}^{-1}$)’

53. Page 1748, Line 8: “This paper describes a [a coupled vadose zone – Groundwater] model for pesticide transport” –

Reply: Thank you. We would insert ‘a coupled vadose zone – groundwater’ in the first sentence of the conclusions section.

54. Page 1748, Line 11: “With the combination of a deep vadose zone and coarse texture, macropore flow was found to be of minor importance for contaminant trans-

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port.” – add that the observed concentrations could be described by the model without assuming macropores. Although a large macrodispersivity was calibrated suggesting deviation from chromatographic transport.

Reply: We would revise the sentence at P1748 L11 according to your suggestion: ‘At regional scale with the combination of a deep vadose zone and coarse texture, the observed concentrations could be described by the model without assuming preferential flow. However, a large macro-dispersivity was calibrated suggesting deviation from chromatographic transport in the vadose zone.’

55. Comment: If macro dispersivity was required, why was the travel time equal to a simple mass balance approach? Furthermore, this approach was not explained with regard to the treatment of surface applied pesticides and how they get to GW.

Reply: We have addressed this in our response to comment no. 5.

56. Page 1748, Line 21: “Further development of the model would require additional field measurements to assess the importance of macropore flow in deep, sandy aquifers. It is also important to characterize the variability of hydraulic conductivity and its effect on contaminant transport in eskers.” Statement 1 is conflicting with what is previously said, namely, that macropore flow does not play a role in deep vadose zones. Statement 2 is not convincing either; what deficiencies in the present study can serve as an argument for studying this?

Reply: We would revise the sentence at P1748 L21-22: ‘Further development of the model would require additional field measurements of macrodispersion to assess its importance in deep, sandy vadose zones.’ Do also read our response to comment no. 8. In the discussions section we would add the following after P1746 L27: ‘In the groundwater model, the heterogeneous properties of glaciofluvial material were not included. According to the performed sensitivity analysis, the hydraulic conductivity turned out to have the highest effect on the model outcome. The variability of hydraulic conductivity may more or less influence the pathways of contaminant transport. If the

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variability is high, the assumption of homogeneous zones within the aquifer might have overestimated the predicted remediation efficiency (Maji & Sudicky, 2008; Rauber et al., 1998).’

57. Table 1: averages for samples taken from 5 locations and depths up to 7 m ? 10 m? I would not use logarithm of K slug test, but the original values, which is consistent with other K units in the rest of the paper.

Reply: We would revise the first sentence in the table text as follows: ‘Analysis of soil samples taken in six, 5 to 10 m deep, soil profiles from the nursery site, including data from Bergvall et al. (2007), and slug tests carried out in the aquifer (Fig .2).’ We would revise the presentation of the K slug tests and present original values in the unit [m d⁻¹].

58. Table 2 with Figure: It is unusual to include a Figure in a Table. The Figure should be stand-alone and larger.

Reply: We would separate the Figure from Table 2.

59. Table 3: Indicate in Table 1 (e.g., using superscript symbols) which quantities were measured, which calibrated, and which represent literature estimates. Add dispersivities to Table 1.

Reply: We would do that.

60. Table 4: obtained for what time frame of simulation?

Reply: We would add the following to the table text: ‘The time step used in Eq. (5) was 180 days, and the time frame was 1st January 1976 to 31st December 2015.’

FIGURE CAPTIONS: Figure A. Observed and simulated water contents in the soil profile that were sampled in 2003. The water content data obtained by the oven-drying method was used to calibrate the vadose zone model. The minimum, mean, and maximum values of water contents were obtained from measurements by the soil moisture

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probe at thirteen occasions (probe, min; mean; max), and was used for validation. The visualized mean of water content data obtained from the model (model, mean) refer to the corresponding thirteen days in the model.

Figure B. Calculated infiltration rates (rainfall + snowmelt – evapotranspiration; left y-axis) and bottom water fluxes at 10.2 m depth (right y-axis). The daily water outflows were generated by the vadose zone model from 1st July 2003 to 30th June 2004. The indicated date of calibration refers to 1st November 2003.

Figure C. Simulated and analyzed concentrations of dichlobenil at one identified hotspot (Fig. 2) in the vadose zone of the nursery field. The detection limit (d.l.) of the analyzed samples was about $10 \mu\text{g l}^{-1}$. The date of calibration was 1st November 2003. The assumed application rate of dichlobenil was 1.7 g m^{-2} , applied at 24 times between 1976 and 1991.

Figure D. Simulated mass fluxes (left y-axis) and concentrations (right y-axis) of BAM at the bottom of the 10.2 m deep vadose zone model. The daily outflows and concentrations of BAM were generated by the vadose zone model from 1st July 2003 to 30th June 2004. The assumed application rate of dichlobenil was 0.17 g m^{-2} , which was ten times lower than the rate in the calibrated model. The BAM concentrations from the vadose zone model were used as input to the groundwater model.

This concludes our response to the comments from Dr. habil. Köhne. We believe that the manuscript has improved significantly during this progress, but we are happy to respond to any other concerns that may arise.

Sincerely, Martin Bergvall (on behalf of all the authors)

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 8, 1729, 2011.

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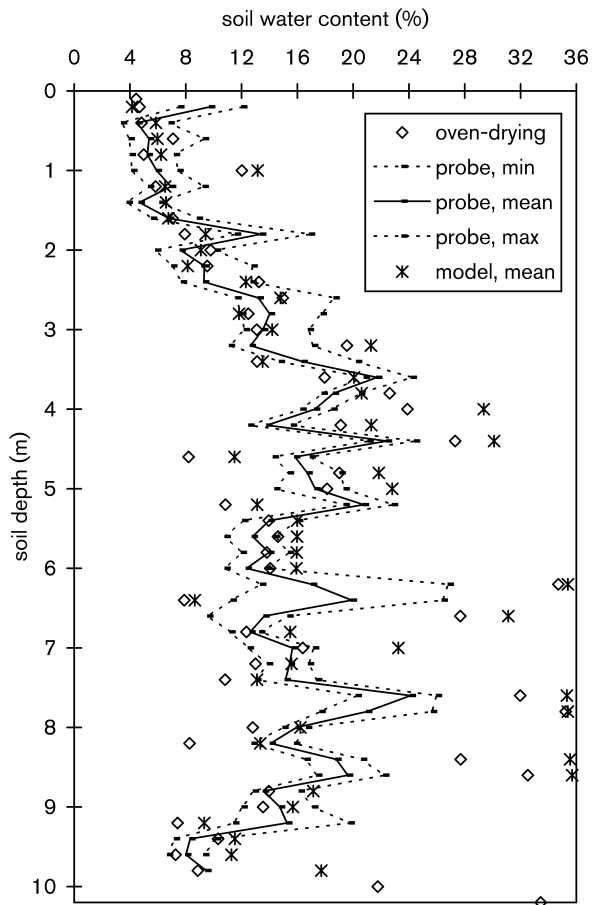


Fig. 1. Figure A

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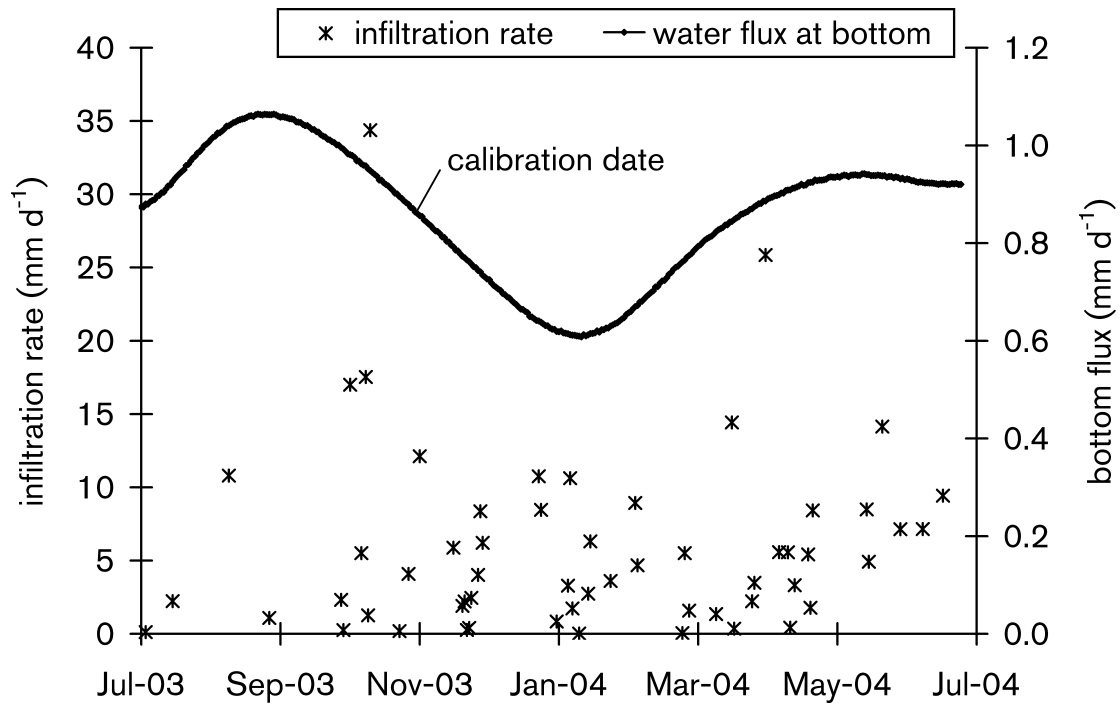


Fig. 2. Figure B

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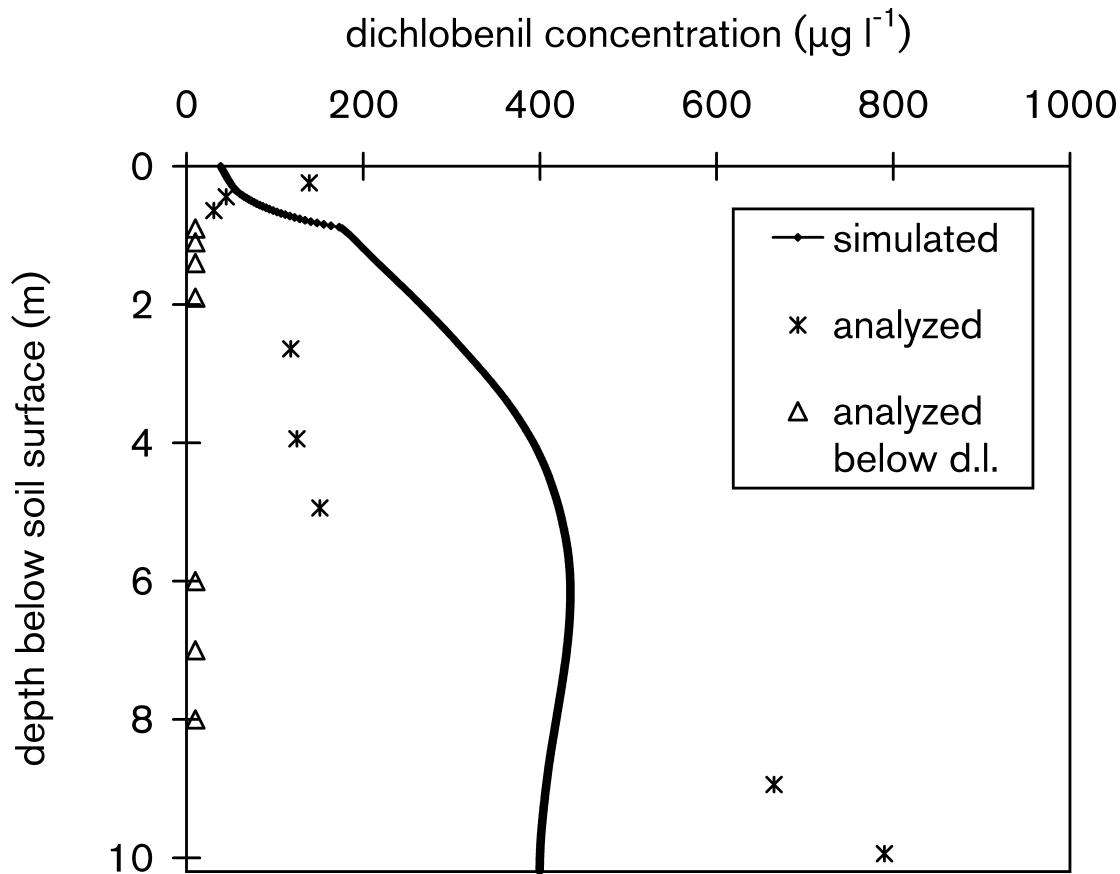


Fig. 3. Figure C

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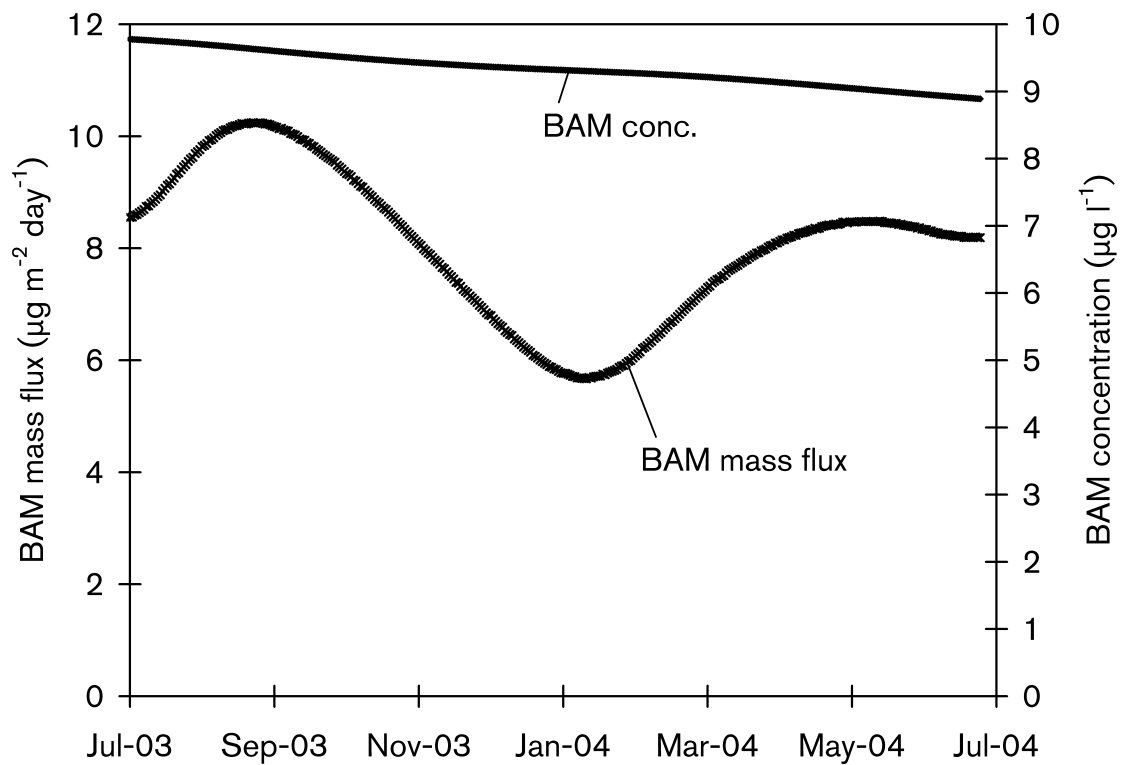


Fig. 4. Figure D

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