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Dilution of pesticides in groundwater during advective dispersive transport

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ABSTRACT

The impact of dispersion on the concentration of pesticide in the saturated groundwater was studied using scenario-type computer simulations for an area in the Netherlands (Lochem). The simulations demonstrated concentrations decrease to with depth; however, the rate of reduction is not systematic and varies throughout the area. At the boundaries between maize land and grassland the reduction is considerable. In other parts of the area,), the reduction is limited and the concentrations gradually approach input concentrations, especially for longterm applications (i.e. > 10 years.

The decrease in concentration by dispersion is based on the mixing of contaminated water with water from the surrounding areas. Therefore the reduction for diffuse contamination or in cases where the load is maintained at a certain level for a long period of time, will be low. The effect of dispersion is greatest in leveling out peaks. The final rate of dilution depends highly on the situation in the surrounding area. Occurrence of dispersion itself is no guarantee that concentrations at a depth of 10 m will be reduced to below the threshold level when they are above the threshold level in the uppermost groundwater.

SUMMARY

This report addresses the question of whether dilution by dispersion contributes substantially in reducing the concentrations of pesticides in the saturated zone. In Dutch legislation on registration of pesticides it is mentioned that in the uppermost groundwater concentrations above the threshold value of 0.1 μg/l are allowed, if it can be demonstrated that, due to transformation processes, the concentration drops to below the threshold value at a depth of 10 m below soil surface. Dispersion is not mentioned explicitly, although this process also may reduce the concentration.

The impact of dispersion on the concentration of pesticide in saturated groundwater was investigated using scenario-type computer simulations for an area in the eastern part of the Netherlands (Lochem). A realistic load of pesticides was assumed at the phreatic surface, below arable land. The spatial variation of the load was related to the distribution of maize-land. Field data were used for the rate of groundwater recharge. To focus attention on the effect of dispersion, other processes like bio-transformation and sorption were left out of the model.

The simulations demonstrated concentrations to decrease with depth. However, the rate of reduction is not systematic and varies throughout the area. On the boundaries between maize-land and grassland the reduction in concentration is considerable. In other parts of the area the reduction is limited and the concentrations gradually approach the input concentrations, especially for long term applications (i.e. > 10 years). After a simulation period of 60 years the decrease is only a few per cent in some places. The effect of dispersion on periodic fluctuations was also investigated in a crop rotation scenario, where pesticides were applied for one year in a four-year period. The results indicate a considerably attenuation of the peaks, but the fluctuations were still noticeable.

The decrease in concentration by dispersion is based on the mixing of contaminated water with water from the surrounding areas. Therefore, the reduction in concentration will be low when the contamination is diffuse or when the load is maintained at a certain level for a long period of time. The effect of dispersion is most important in leveling out peaks, both in time and in space. The final rate of dilution depends highly on the situation in the surrounding area. Occurrence of dispersion itself is not a guarantee that concentrations at a depth of 10 m will be reduced to below the critical level when they are above the critical level in the uppermost groundwater.

Keywords: pesticides, solute transport, saturated groundwater, simulation, scenarios.

SAMENVATTING

Dit rapport gaat in op de vraag of verdunning door dispersie een belangrijk aandeel levert in de reductie van de pesticide concentraties in de verzadigde zone. In de Nederlandse wetgeving over de toelating van pesticiden wordt vermeld dat in het bovenste grondwater concentraties boven de drempelwaarde van 0.1 µg/l zijn toegestaan, mits kan worden aangetoond dat door transformatieprocessen de concentratie op 10 meter diepte is gedaald tot onder de drempelwaarde. Dispersie wordt niet met name genoemd, ofschoon dit proces eveneens de concentraties kan reduceren.

In dit rapport wordt de invloed van dispersie op de pesticideconcentratie in het verzadigde grondwater nader onderzocht aan de hand van scenario-georiënteerde computer-simulaties voor een gebied in het oosten van Nederland (Lochem). In gebieden met bouwland werd aan het freatisch oppervlak een realistische belasting van pesticide verondersteld. De ruimtelijke variatie van de belasting werd gerelateerd aan de verdeling van maisland. Voor de nuttige neerslag werden veldgegevens gebruikt. Om de aandacht te concentreren op dispersie, werden processen als biotransformatie en sorptie niet in het model opgenomen.

De simulaties tonen aan dat de concentratie afneemt met de diepte. De mate waarin dit gebeurt is echter niet systematisch en varieert over het gebied. Aan de randen, waar maisland en grasland aan elkaar grenzen, is de reductie aanzienlijk. In andere delen, vooral bij lange belastingperioden, is de reductie beperkt en naderen de concentraties de ingangsconcentraties. Na een simulatie periode van 60 jaar is op sommige plaatsen de afname slechts een paar procent. Het effect van dispersie werd ook onderzocht voor een rotatiescenario, waar pesticiden een jaar lang worden toegepast in een periode van 4 jaar. De resultaten laten zien dat periodieke fluctuaties aanzienlijk worden afgevlakt, maar ze blijven herkenbaar.

De afname in concentratie door dispersie is gebaseerd op menging van verontreinigd water met water uit de omringende gebieden. Het is duidelijk dat de reductie klein zal zijn, als er sprake is van een diffuse verontreiniging, of wanneer de belasting lange tijd op een bepaald niveau blijft. Het bealngrijkste effect van dispersie is het afvlakken van pieken, zowel ruimtelijk als in de tijd. De uiteindelijke verdunning hangt in hoge mate af van hetgeen er in de omgeving aan de hand is. Het optreden van dispersie is daarom op zich geen garantie dat concentraties op 10 meter diepte worden gereduceerd tot beneden de drempelwaarde, wanneer ze in het bovenste grondwater boven de kritische waarde liggen.

Trefwoorden: bestrijdingsmiddelen, transport van opgeloste stoffen, verzadigd grondwater, simulatie, scenario's.

1 INTRODUCTION

Registration of pesticides is regulated in a number of national and international laws; amongst others the EC directive 91/414 and the Dutch Pesticide Act of 1962 and their amendments. The Netherlands has specified several environmental criteria in the so-called AMvB 3a of January 23rd, 1995, specifically with respect to 'persistence in the soil', 'leaching to groundwater' and 'risks to aquatic organisms'. Article 6 of this AMvB specifies that a compound shall not be registered, if in the uppermost groundwater the measured or calculated concentration of the compound or its metabolites is above the threshold level of 0.1 μg/l, unless it is demonstrated that, due to transformation processes, the concentration will decline to levels below the threshold level and that the water will fulfill the criterion at a depth of 10 m below soil surface.

As stated above, the AMvB mentions decline of concentration only by transformation processes. Several people, however, stated that physical processes as dilution and hydrodynamic dispersion can lead also to a decline of concentrations down to a depth of 10 m and, therefore, may contribute in reaching the required quality criterion. Information on dispersion , however, was too little to be included in the legislation. Therefore, it was decided to investigate the impact of dispersion on concentrations of pesticides in groundwater.

The purpose of the present investigation is to determine to what extent dispersion can affect the pesticide concentration in the saturated zone under realistic agricultural practices. Special attention is paid to:

- concentrations at a depth level of 10 m below groundwater table
- concentrations in water abstraction wells.

On the basis of this investigation inclusion of dispersion and dilution processes in the evaluation of the registration of pesticides can be considered.

The advisory committee of the investigation specifically asked to consider the following situations and items:

1. upward and downward seepage;
2. natural situations and situations influenced by abstraction practices;
3. continuous and intermittent use;
4. the equilibrium situation as well as the dynamic situations towards the equilibrium.

Given the purpose of the study and the specific requirements of the advisory committee, it was decided to carry out an investigation in a small, rather well-known area in the neighborhood of the drinking water pumping station Lochem, located in the eastern part of the Netherlands.. The hydrological situation around this station has been studied for several years (Kovar et al., 1996). The hydrological data used in this report are identical to those used in earlier studies for the Dutch Ministry of Housing, Spatial Planning and the Environment. The hydrological model LGM (National Groundwater Model) has been applied, while the solute transport is simulated by the module LGMCAD (Uffink, 1996). The hydrological model uses steady flow conditions, resulting in stationary groundwater flow. Solute transport, however, is simulated in a non-stationary manner, taking temporal variations of the input of solutes into account. With respect to the type of pesticide, the study is generic, i.e. no specific properties of a pesticide have been considered. Transport processes have been simplified to advection-dispersion, in order to focus attention on the physical dilution process. Absorption/desorption and biodegradation (decay) are not included, also to avoid complexity, to simplify data collection and to reduce computer execution time.

The spatial distribution of the leaching of pesticides is based on the present agricultural use of the soil combined with results of simulations in the unsaturated zone, carried out for various soil types (Tiktak, et al., 1996). Temporal variation in the application of pesticides have been taken into account by considering several scenarios.

Chapters 2 and 3 describe the study area in detail. Chapter 4 presents the scenarios developed for studying various aspects, while in chapter 5 the results are presented and discussed. Finally, in chapter 6 conclusions are given.

2 SITE-DESCRIPTION

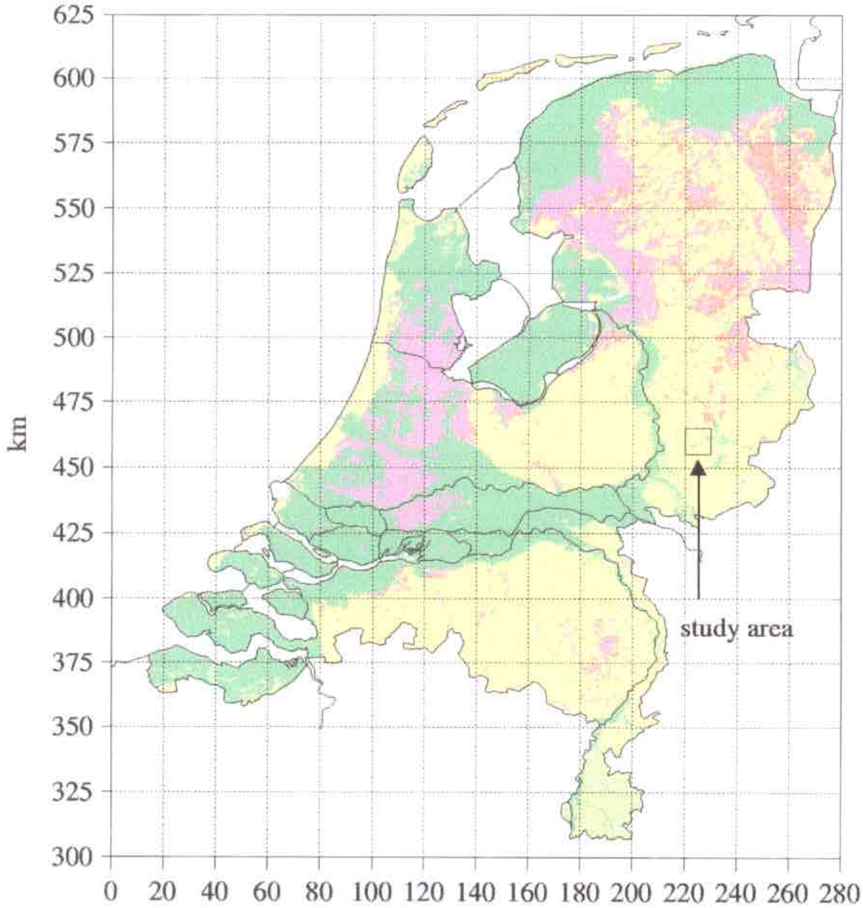


Figure 1 Location of study area 'Lochem'

The study region is located in the eastern part of the Netherlands (figure 1) and covers an area of 10 X 10 km^2. It is referred to as the 'Lochem' area, after the main city in the region. The non-urbanized part consists mainly of grass-land, alternated by maize-land. A detailed map (figure 2) shows the lands cultivated with maize. The map also indicates the groundwater recharge areas and the areas where, due to the detailed drainage system, no groundwater recharge occurs (gray colored).

The main courses of surface water are indicated in blue. In the northern part of the area runs the 'Twente-canal' and a creek called the 'Berkel', while in the South the 'Veengoot' is located. All these streams run from East to West. A few contour lines in the gray zone (drainage zone) indicate relatively high rates of drainage along the Twente-canal and in the south along the 'Veengoot'. Along the Berkel both seepage and infiltration occurs. This means, in principle, that pesticides drained in the upstream part of the Berkel may re-enter the groundwater system further downstream. This source of pesticides, however, has not been taken into account in the model.

The city of Lochem is located in the northern part of the region. Close to the city is a

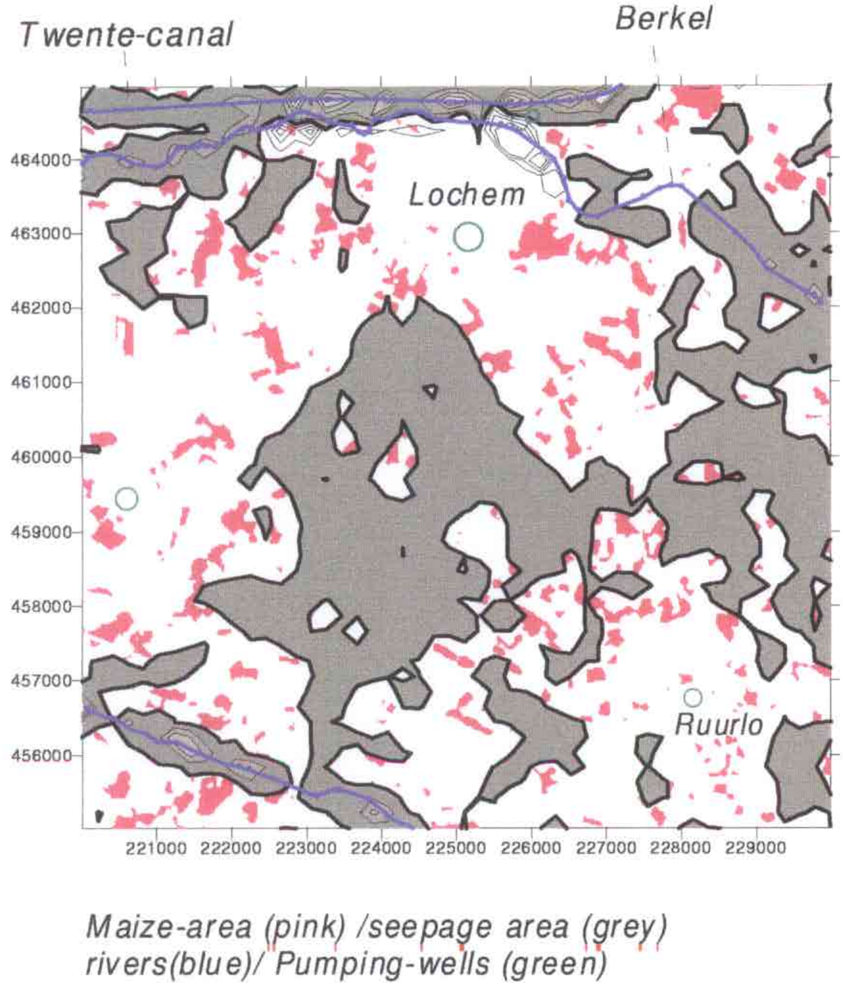


Figure 2 Land-use and seepage area

groundwater pumping station with the same name. The pumping station extracts water for the public drinking water supply with a yearly capacity of 2 million m^3. In the south-east of the area lies the village of Ruurlo, also with a small drinking water pumping station in its vicinity. A third pumping well is located in the western part of the study area.

Figure 3 gives the elevation of the terrain in meters above standard datum level (NAP). The overall relief shows a downward trend in the direction South-East to North-West, except South-East of the city of Lochem, where the surface is more elevated than in the rest of the area. Three hills can be distinguished: the 'Paaschberg', the 'Lochemerberg' and the 'Kalenberg'. In these higher parts groundwater recharge is positive (infiltration). The groundwater pumping station Lochem is located on the Paaschberg.

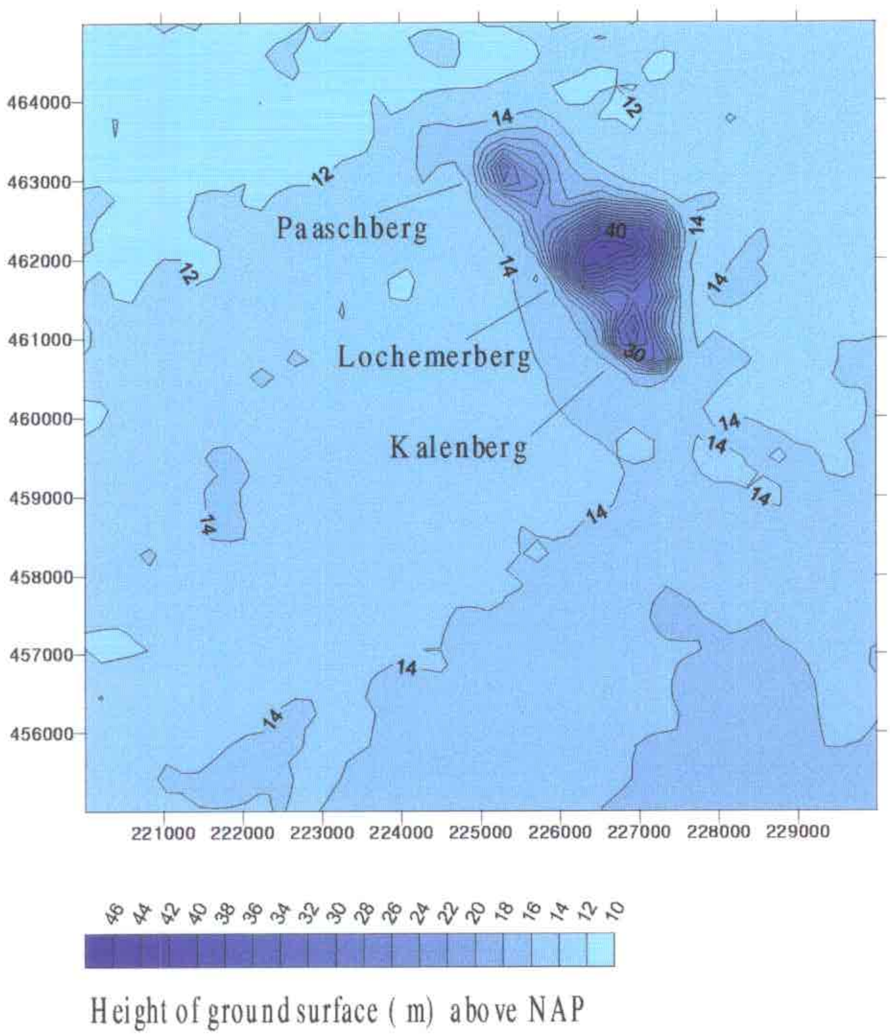


Figure 3 Contour map of ground surface

3 GEOHYDROLOGICAL SCHEMATIZATION

The geohydrological system is simplified to one single unconfined (phreatic) aquifer with an averaged thickness of 60 m. The height of the phreatic surface (groundwater table) varies from 10 to 16 meter above NAP. The prevailing gradient is from South-East to North-West (figure 4). The direction and magnitude of the horizontal component of the groundwater flow is given in figure 5, also showing the prevailing South-East to North-West direction. Exceptions occur near the pumping wells, where locally high groundwater velocities are found and along the northern boundary of the area, where high velocities are due to presence of the 'Twente-canal'.

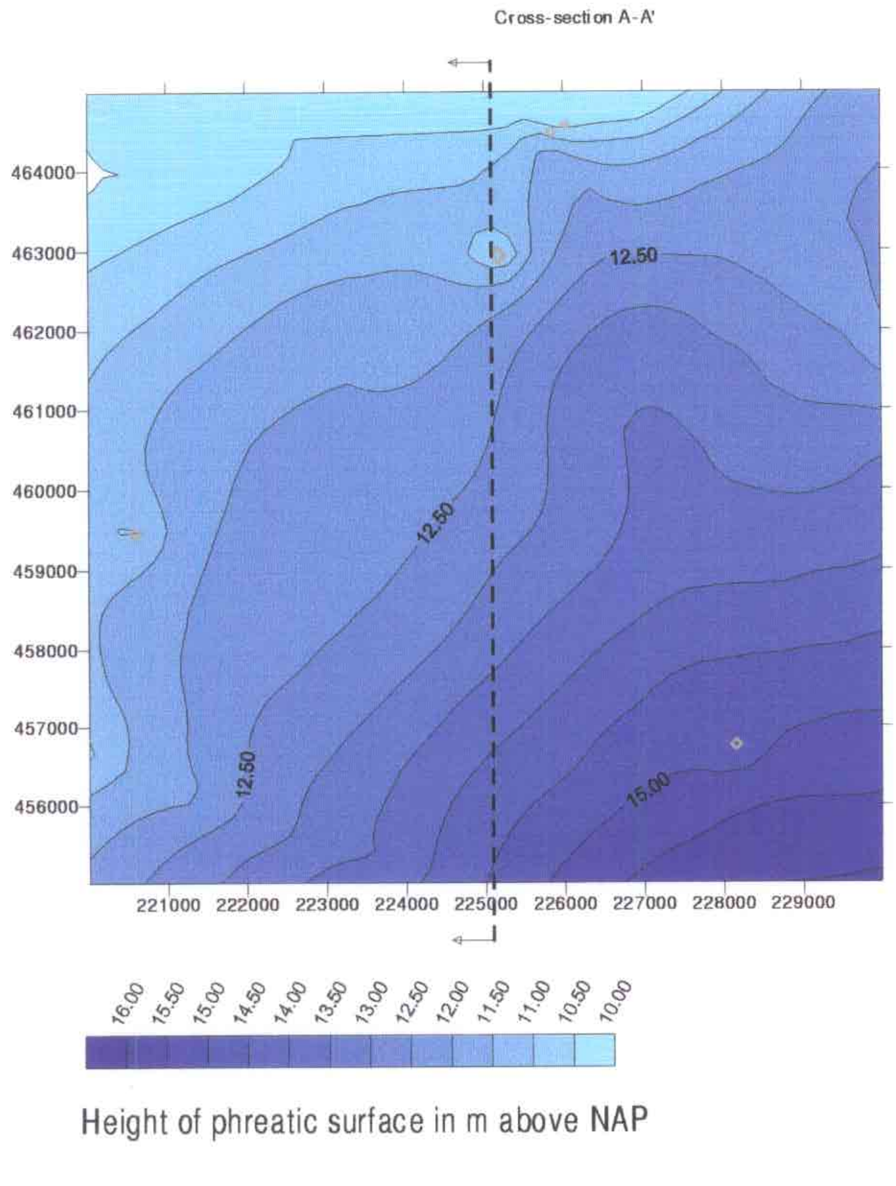


Figure 4 Phreatic surface in the study area

Another typical feature that is recognized is the infiltration of groundwater at the 'Lochemer berg'. Following a more or less radial flow pattern the water moves away from the center of infiltration, which is located at the top of the 'Lochemer berg'. The magnitude of the groundwater flow varies from zero to about 0.16 m/day. When the extreme velocities due to the pumping wells are left out of consideration, an average velocity of 0.04 m/day is found. Note that the arrows are positioned in such a way that they represent the velocity at the point halfway the head and tail of the arrow. The length of the arrow corresponds with an advective displacement over a period of 10000 days (30 years).

A cross-section in the direction South-North, running over the pumping well of the station Lochem (indicated in figure 4 as A-A') is given in figure 6. In this figure the pumping station is schematized as a single well with an average discharge of 5133 m^3 / day. The impervious base of the aquifer is found at a depth varying from 40 m -NAP in the South to 60 m -NAP in the North.

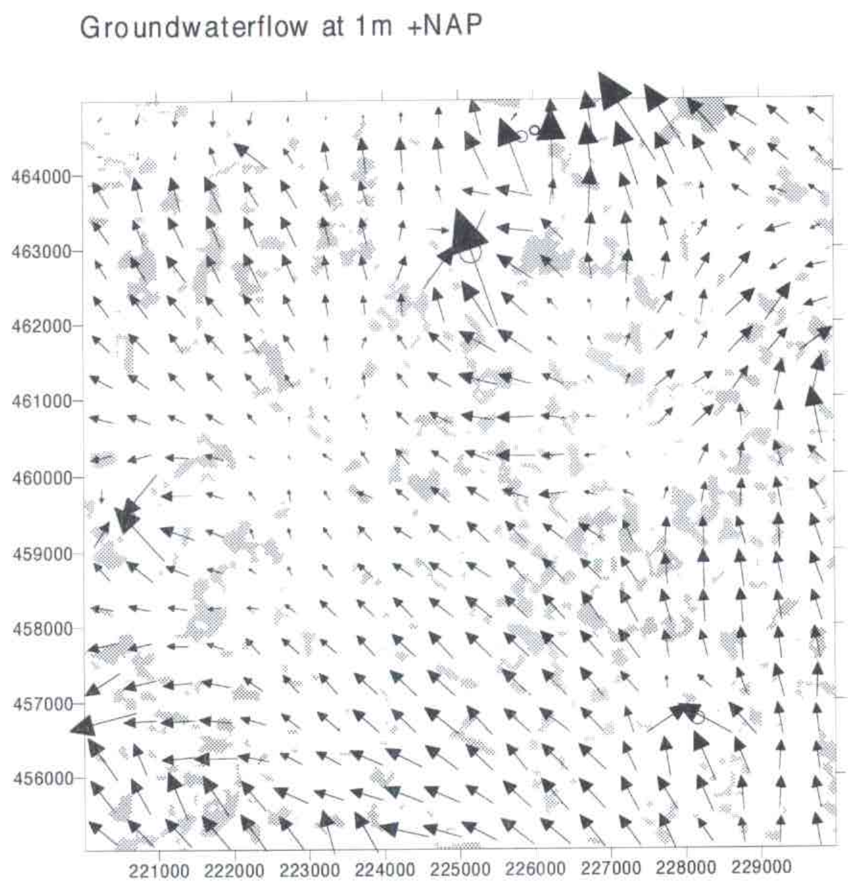


Figure 5 Magnitude and direction of the horizontal groundwater flow.

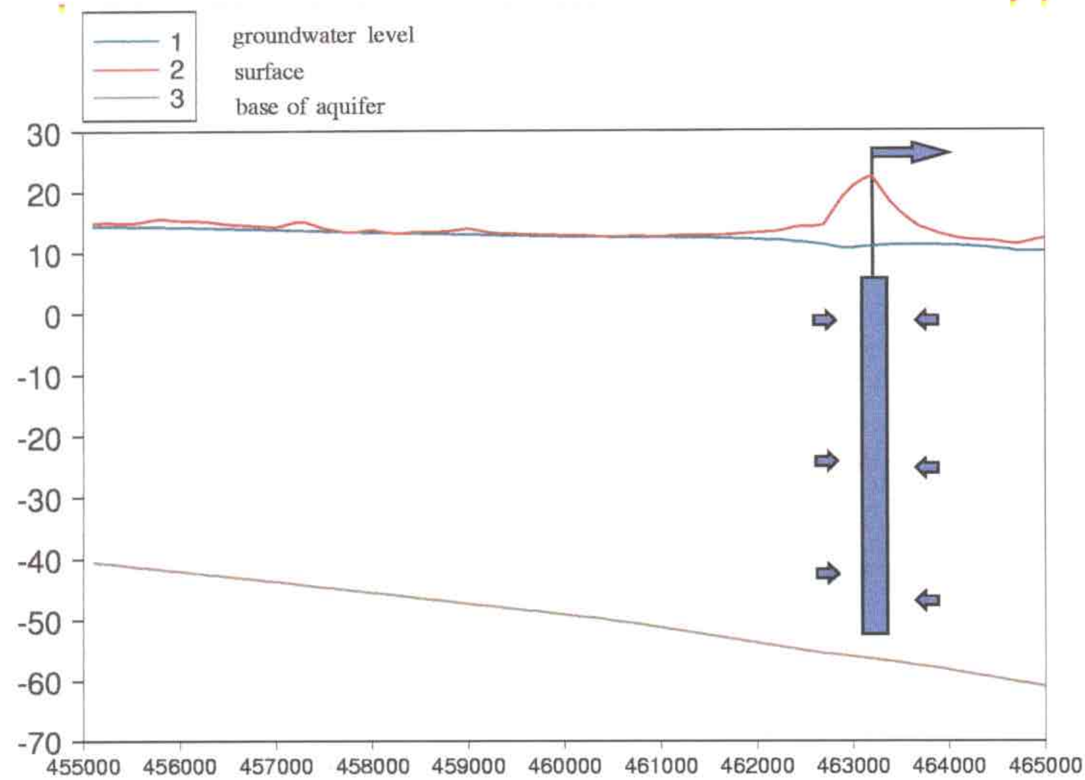


Figure 6 Cross-section A-A' as shown on figure 4. Vertical axis elevation in meter above NAP. Horizontal axis y-coordinate along the cross-section

3.1 Groundwater Recharge and Leaching rates of pesticides

The natural groundwater recharge in the area varies both in space and in time. Since the hydrological model is based on stationary flow, temporal variations in the recharge quantities are not considered. The pesticides examined in this study originate from lands with maize (corn) cultivation. Thus, the leaching rate depends directly on the distribution of maize-cultivated lands, as shown by the pink areas in figure 2. In the gray marked areas of figure 2, the drainage system prevents groundwater recharge and, consequently, here leaching of pesticides is zero.

Although the groundwater flow model is stationary, temporal variations in the pesticide concentration of the recharge have been taken into account by considering various leaching patterns. These patterns, defined in several scenarios, are described in chapter 4. The pesticide concentration of the recharge is calculated as follows. On a unit area of maize land [m^2] an amount m [mg] leaches to the groundwater system during 1 year. It is assumed that this amount does not depend on the rate of recharge at that particular parcel. If the recharge rate is denoted by q [m/day], then the concentration follows from $c = m/(365\ q)$. For m is taken a value of 0.1 mg/m^2. (Tiktak et al., 1996) Figures 7a-7d illustrate several steps during the procedure. Figure 7a gives the distribution of the natural groundwater recharge over the total area. In the white-colored areas no groundwater recharge occurs, so no pesticides can reach the groundwater. Figure 7b gives the distribution of maize land. A combination of 6a and 6b, yields the rate of recharge on maize land, (figure 7c). The white areas in figure 7c correspond to areas that are white in 7a or 7b. Figure 7d is surface plot based for the same data as for 7c. The areas where due to the drainage system no groundwater recharge occurs, no input of pesticides is applied. The final pesticide concentration in the groundwater recharge is given in figure 7e, using the same color-scale as used for the simulation results.

The leaching data are available with a resolution of 25 X 25 m^2, in a grid format consisting of 400 X 400 grid cells, with a total grid dimension of 10 X 10 km^2. The total area of the maize land is 14.048 10^6 m^2 (14 % of total), while the area of maize land situated in the groundwater recharge zone is 8.3906 10^6 m^2. Consequently, the total amount of pesticides leaching to the groundwater in this area is 839.06 gram per year.

3.2 Other data

The porosity of the aquifer is 0.3. Dispersion is simulated with a longitudinal dispersivity α_L of 10 m and a transverse dispersivity α_T of 2 m. These values are estimated, since more exact data for the dispersivity in the study area are not available. With respect to the dispersivity it is noted that this parameter not only represents a physical phenomenon, but also depends on the scale of the hydrological model. In the latter case it is a measure for the *macrodispersion* and its purpose is to describe the effect of small-scale heterogeneities. Small-scale here means smaller than the size of the elements of the hydrological model, which in the present study was 250m. This value may be considered to be an upper limit for α_L. As a lower limit data from small-scale field test may be considered. For a sandy aquifer along the coast in the Netherlands values varying from 10^{-2} m to 3 m have been found for α_L (Uffink, 1990). The value chosen for this study (α_L = 10m) falls inside these limits.

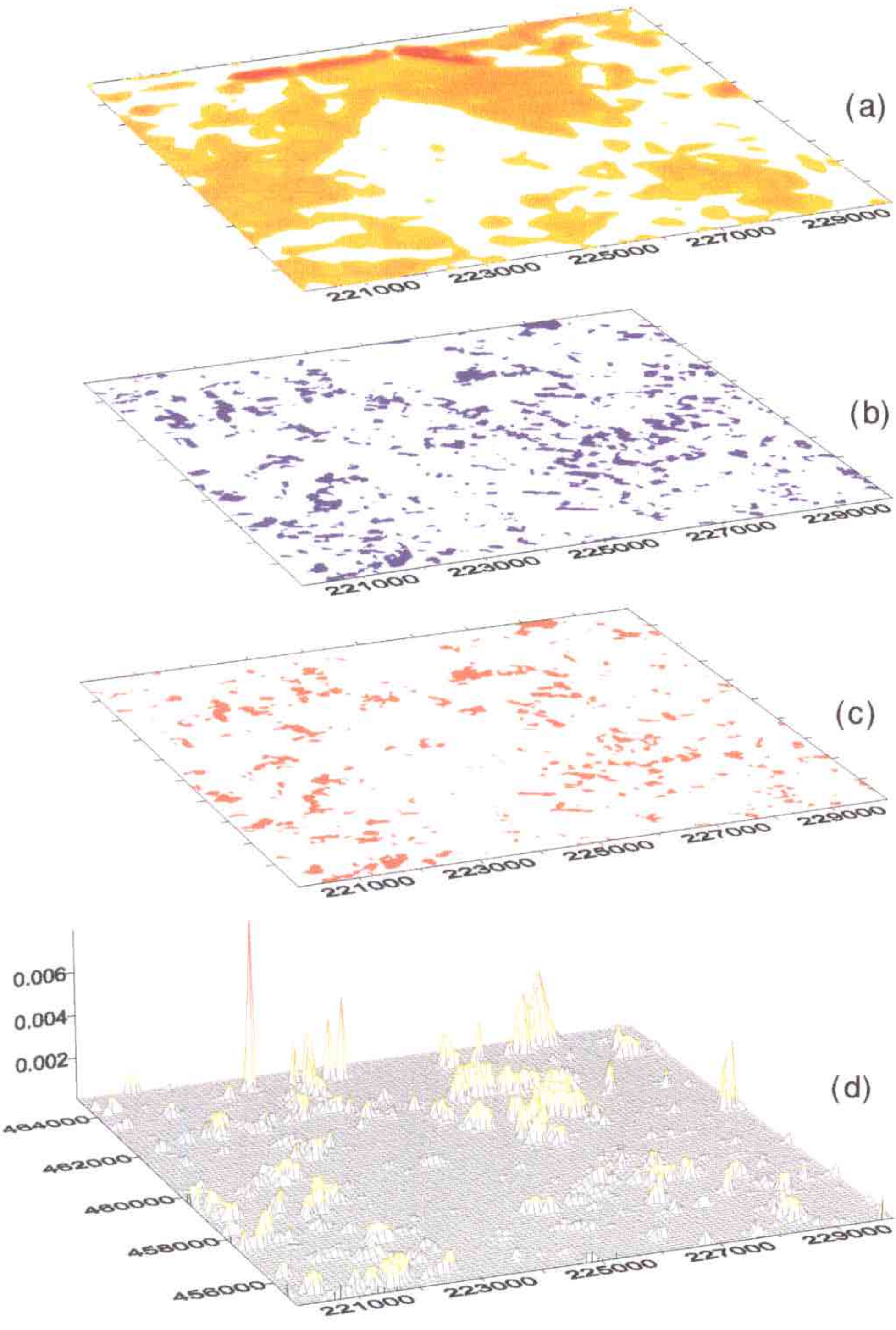


Figure 7 (a) Distribution of natural groundwater recharge; (b) distribution of maize-land; (c) and (d) combination of (a) and (b). Vertical scale in (d) gives groundwater recharge in m/day.

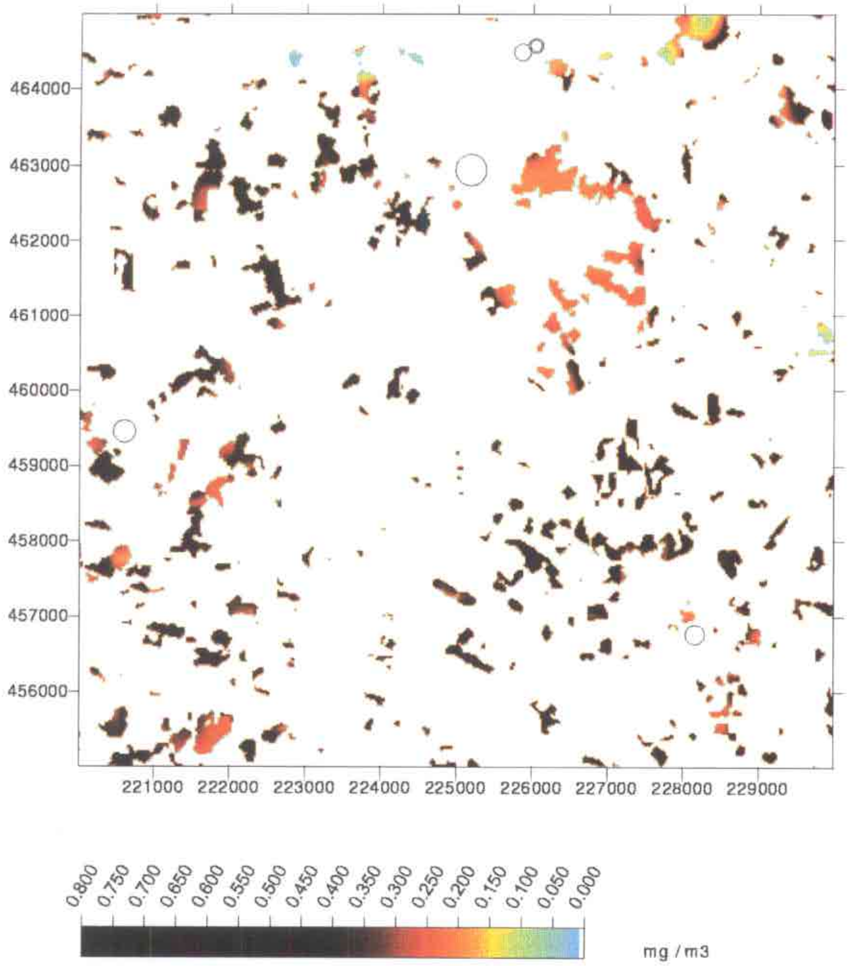


Figure 7 (e). Concentration of pesticide in the groundwater recharge

4 SCENARIOS

With respect to the distribution in time several scenarios are considered. A brief description of the scenarios is given below.

4.1 Basic scenario

For the temporal fluctuation of the leaching process, a period of 10 years has been considered. This period corresponds with the time period generally mentioned in registrations of pesticides. It should be noted that after this 10-year interval the quantity of groundwater recharge does not change. It only no longer contains pesticides.

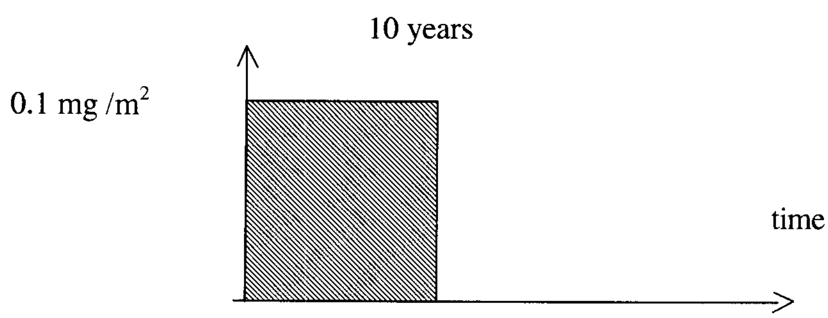


Figure 8 Leaching pattern in time for pesticides in basic scenario

4.2 Additional scenario 1

The simulation data in this scenario are the same as in the basic scenario, but additional output data is produced. Spatial distribution will be considered (both at 10 m and at 20 m depth) for the moments 10, 20, 30, 40 years after the start of the pesticide-application. Also more breakthrough data will be presented.

4.3 Scenario 2 (continuous leaching)

Same simulation parameters as in the basic scenario except for the leaching period. Instead of a 10-year leaching period a continuous leaching is assumed. The output is like in the scenario 1

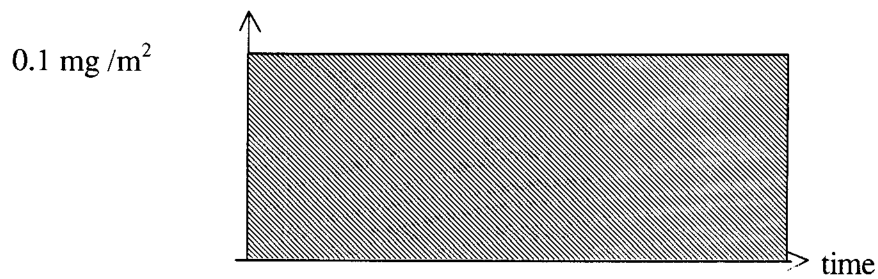


Figure 9 Leaching pattern for scenario 2

4.4 Scenario 3 (discontinuous leaching)

Same simulation data as the basic scenario except for the leaching pattern. Here the dose of pesticides is supposed to be given during one year, followed by three years with no dose (see figure 10).

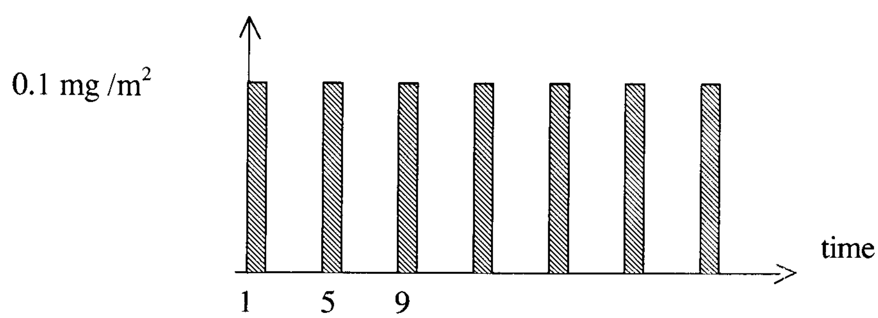


Figure 10 Leaching pattern for scenario 3

5 RESULTS

5.1 Basic scenario

The output focuses on:

- The spatial distribution of the concentration at a depth of 10 meter under the groundwater table and at a time 4 years after the application of the pesticides has been stopped (also see fig. 7)
- The spatial distribution of the concentration at a depth of 20 meter under the phreatic head and at the same time as mentioned above
- A breakthrough curve of the pumping well at PS Lochem.

Figures 11 shows the spatial concentration distribution at a time 14 years after the start of the application (4 year after the application had been stopped). In figure 11a the concentration is presented at a depth of 10 meters below the groundwater surface, while figure 11b gives the distribution at 20 meter below groundwater. It appears that at 10 meter depth the distribution is not much different from the distribution at 20 meter. However, one may not conclude from this that dilution does not vary with depth. In this particular situation, during a period of 4 years 'clean' water infiltrates at the top that creates dispersive mixing at a lower depth as well. Evidence for this explanation follows from the results of the additional scenario 1.

The relation between the plumes and the location of the maize-land is better seen in figure 12. In this overlay figure 11 (depth of 10 meter) is combined with some elements from figure 2, i.e. the line indicating the separation between seepage area and recharge area (black contour line), and the location of the maize-crop lands (pink-line). The general movement of the plumes is in North-western direction. However, locally the influence of the pumping wells is evident. It is also clear that in the seepage areas no pollution plumes appear.

Figure 13 gives a breakthrough curve for the pumping well at PS Lochem. One can see that the first reaction at the well occurs about 5 years after the leaching process begins. This delay occurs, since the maize areas closest to the well are still about 700 meters away and it takes some years for the pesticides to travel to the well. It is seen also, that the concentration at the well continues to increase after t = 10 years, although the leaching stops. The reason for this is, that pesticides leached at greater distances are still in the groundwater system and considerably more time is required for these solutes to reach the well. Only after 25 years the concentration reaches its peak and then gradually starts decreasing.

Note that in reality:

- the first reaction at the well may take more time than 5 years due to sorption of the pesticide to the soil matrix (for this study sorption to the soil matrix was neglected)
- the concentrations at the well may be lower due to (bio)transformation of the pesticide in the saturated zone (also this process was neglected in this study)
- the time span of the peak observed at the well may either be shorter due to (bio)transformation or longer due to sorption; if both (bio)transformation and sorption occur, it is expected that transformation will dominate the results.

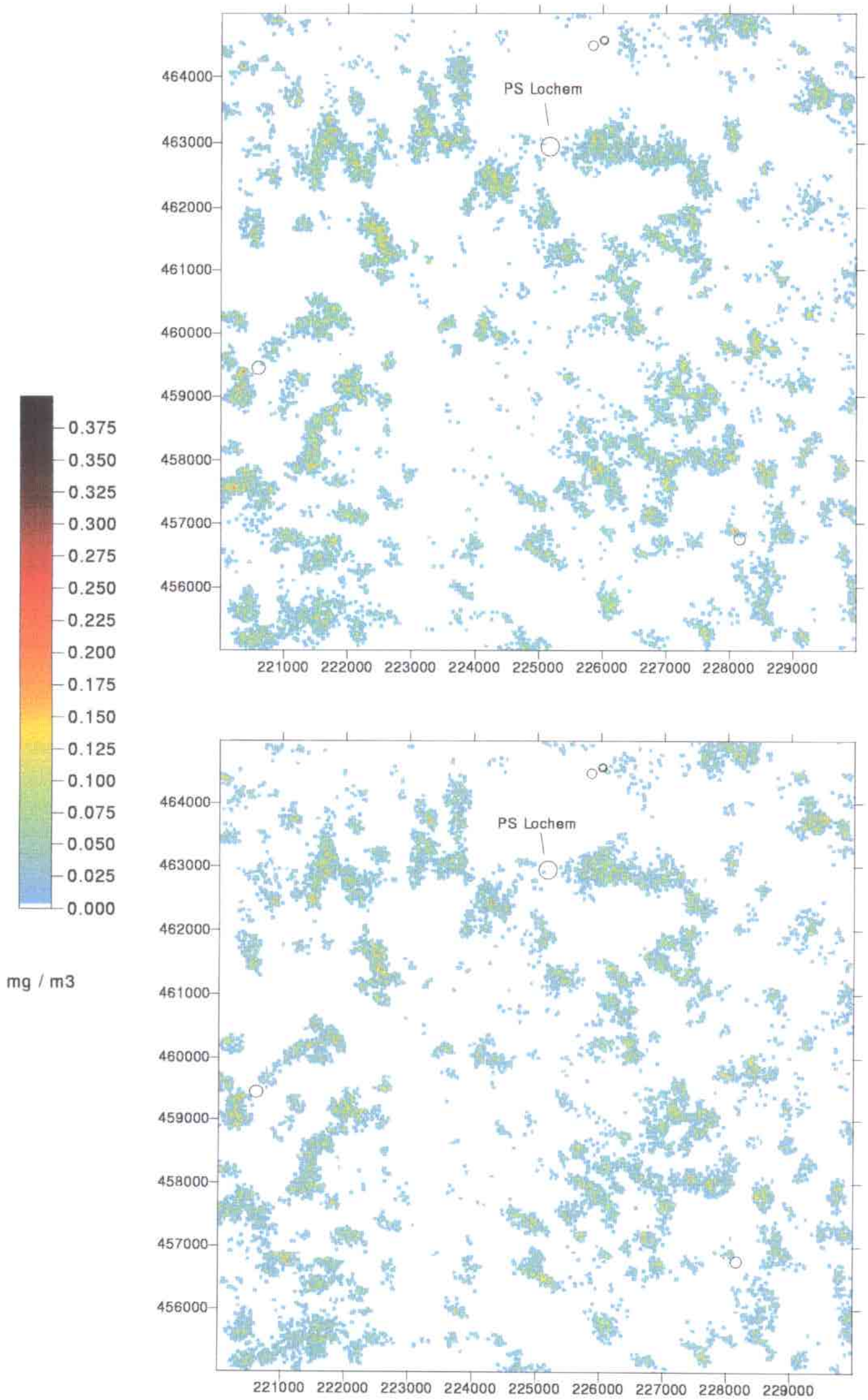


Figure 11 (above) Concentration distribution at t = 14 years and 10 m depth;
(below) Concentration distribution at t = 14 years and 20 m depth

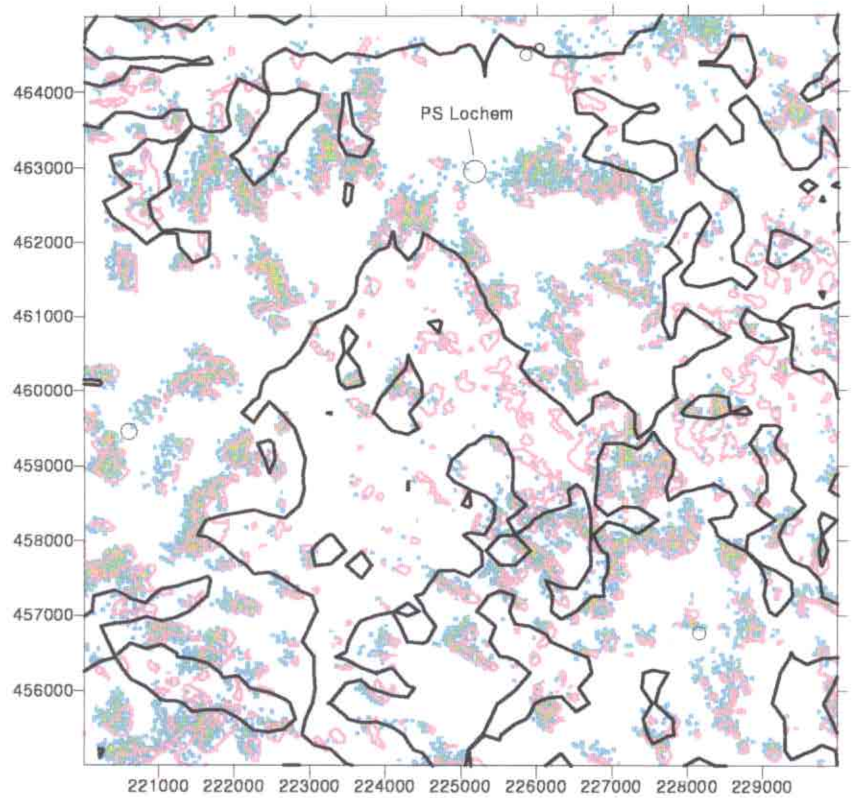


Figure 12 Concentration distribution at t = 14 years and 10 m depth; line separating recharge/seepage (black); maize-land (pink)

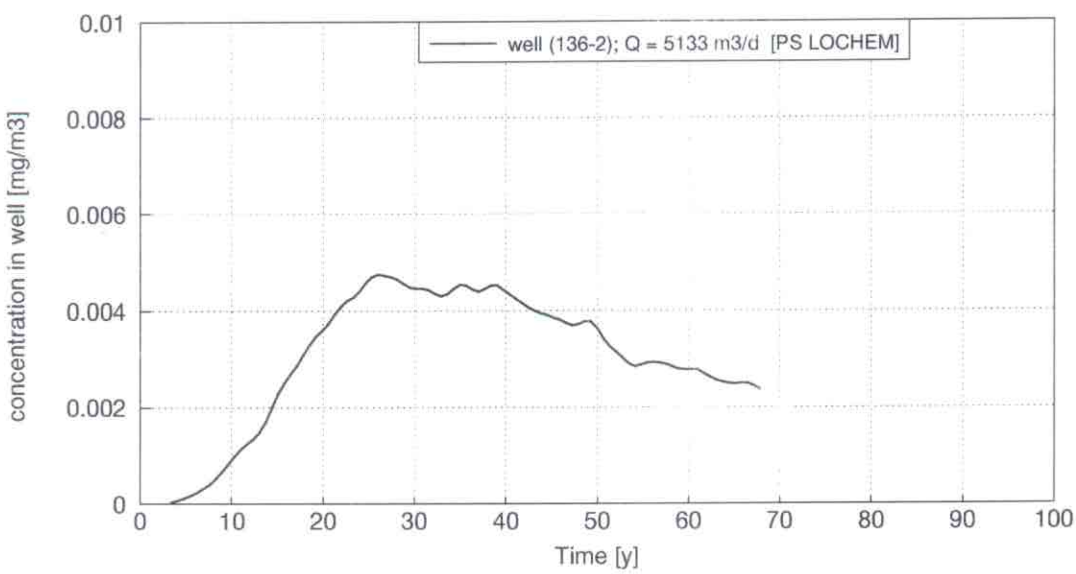


Figure 13 Breakthrough curve at PS Lochem for basic scenario

5.2 Additional scenario 1

Figures 16 and 17 show the concentration distribution at a depth of 10 m below groundwater table at 10, 20, 30 and 40 years after the start of the simulation. The overall concentrations clearly decrease with time. This diluting effect is due to dispersion, since advection causes only a displacement of the solutes without changes in concentration. Besides advection and dispersion no other transport processes have been included in the model. Note that dispersion does not decrease the total amount of pesticides in the groundwater. Lower concentrations occur at the expense of a larger contaminated area.

The figures show a relatively slow displacement of the solutes towards the pumping station. The plumes arrive at the well between $t = 10$ and $t = 20$ years. Another effect of dispersion is that plumes, generated between $t = 0$ and $t = 10$ years, are not fully replaced by clean (i.e. pesticide-free) water during the next interval of 10 years (between $t = 10$ and $t = 20$ years). This would happen in case of purely advective transport. Figure 14 illustrates this effect, which is referred to as 'tailing'. Tailing is evident in the vicinity of the pumping station. The figures 18 and 19 with concentration at a depth of 20 meters below groundwater level display similar features as figures 16 and 17.

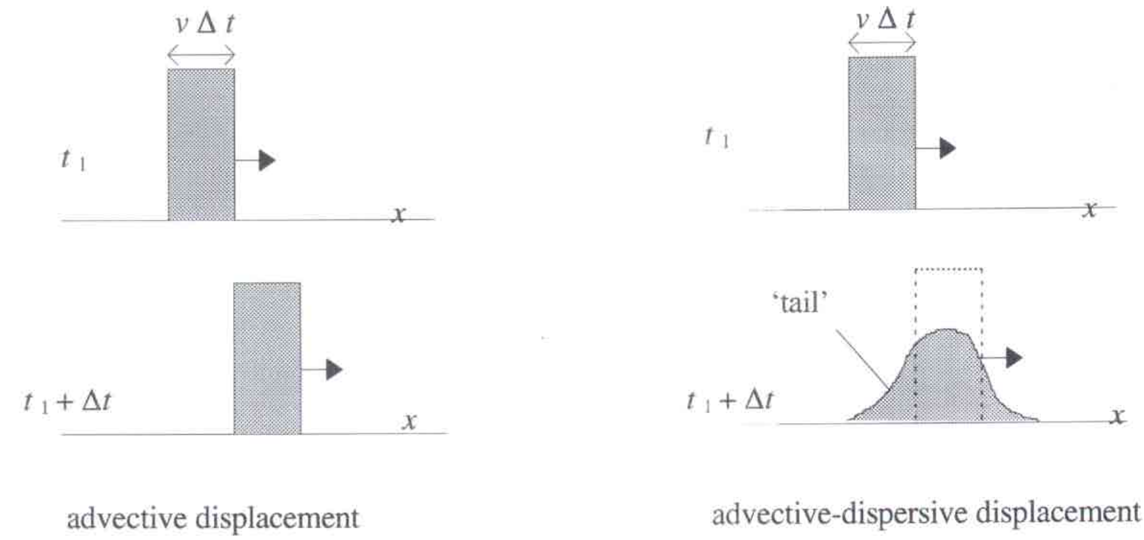


Figure 14 Displacement (with velocity v) of concentration profile. Illustration of 'tailing' effect.

5.3 Cross-section scenario 1

For a vertical cross-section, located along the line *B-B'* in figure 15, the concentration is given for times t equal to 10, 14, 20, 30 and 40 years (figure 20). The tailing effect can be observed here. The solutes are not purely pushed forward by the pesticide-free water that infiltrates after $t = 10$ years. Such a displacement (plug-flow) would be observed if dispersion is absent. Evidently, dispersive mixing occurs, resulting in a gradual decrease of the concentration, and also in a longer residence time of the solutes in the aquifer. This means a longer period for recovery of the groundwater quality. It is noted that at the location of the cross-section the base of the aquifer is located at a depth of around 70 m -NAP. Apparently, the pesticides occur over the total height of the aquifer.

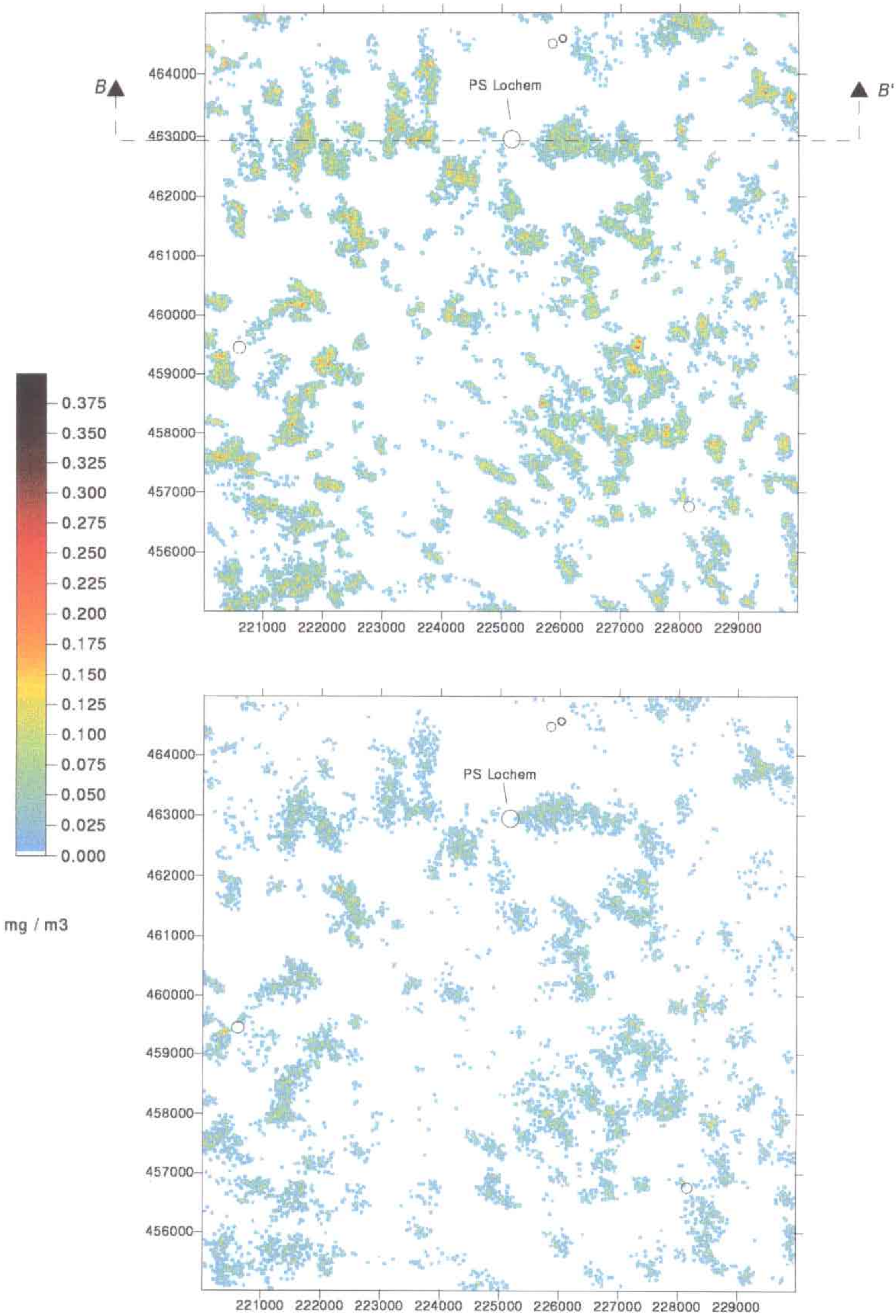


Figure 15 Concentration 10 m below groundwater level at 10 year (above) and 20 year (below)

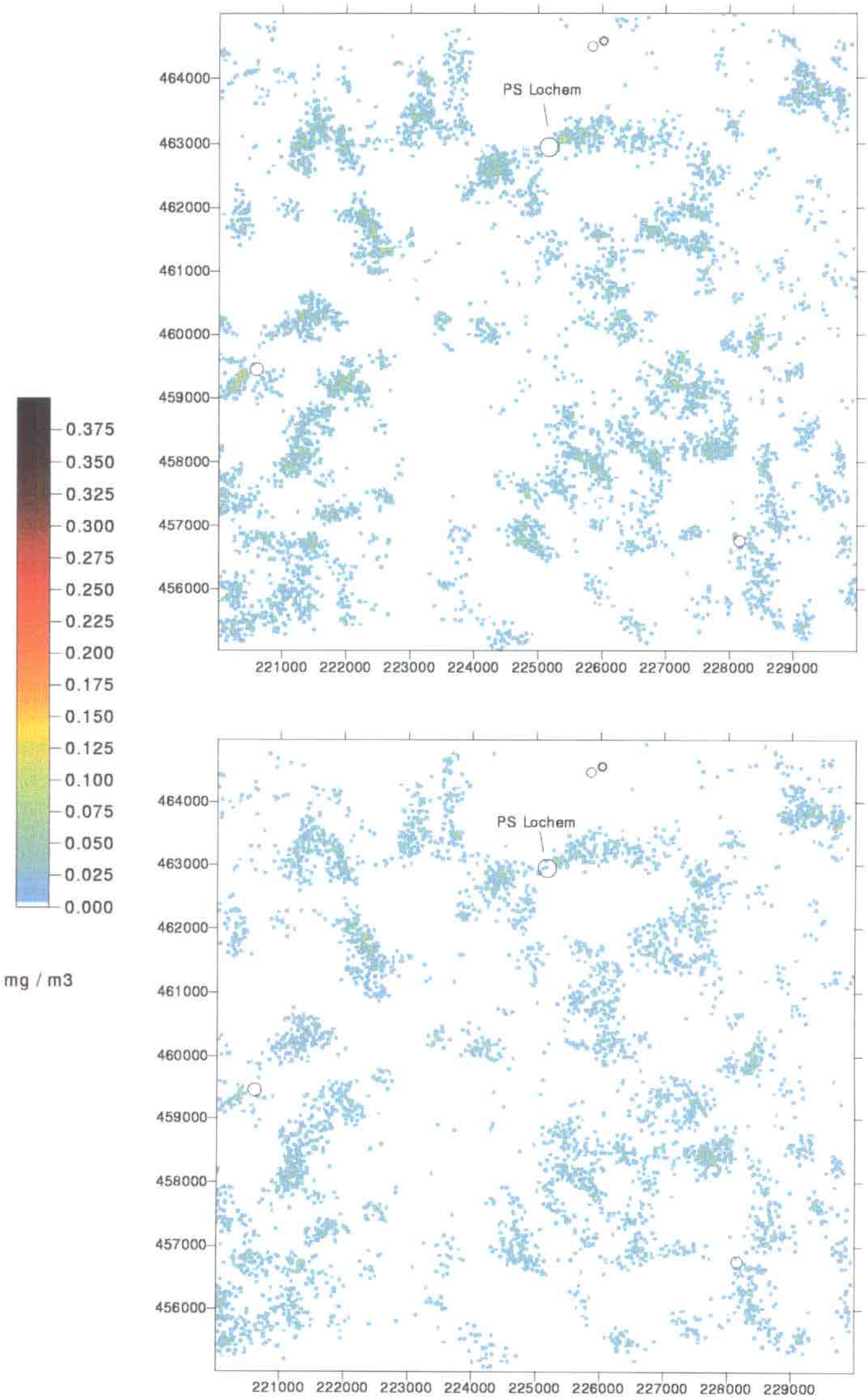


Figure 16 Concentration 10 m below groundwater level at 30 year (above) and 40 year (below)

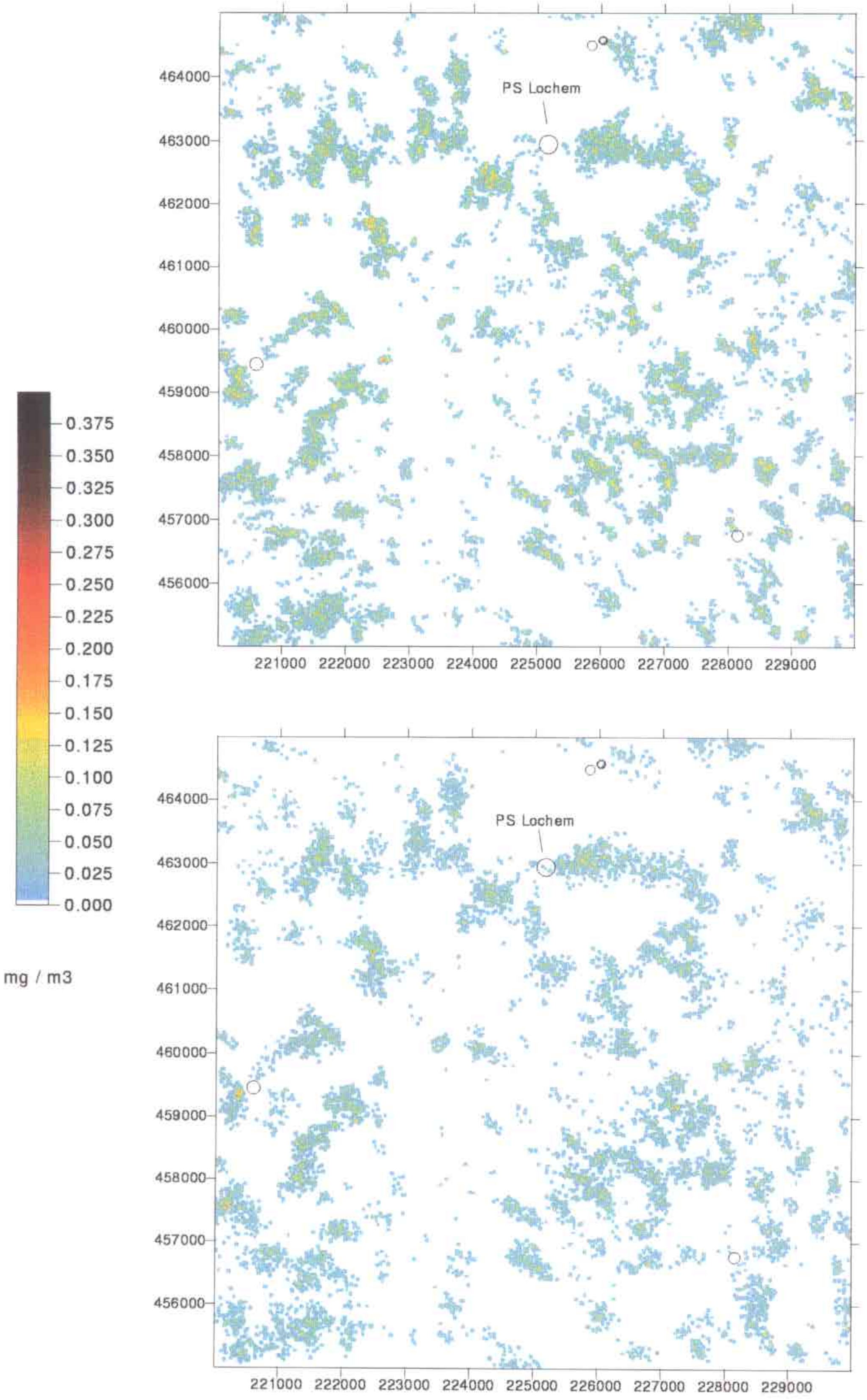


Figure 17 Concentration 20 m below groundwater level at 10 year (above) and 20 year (below)

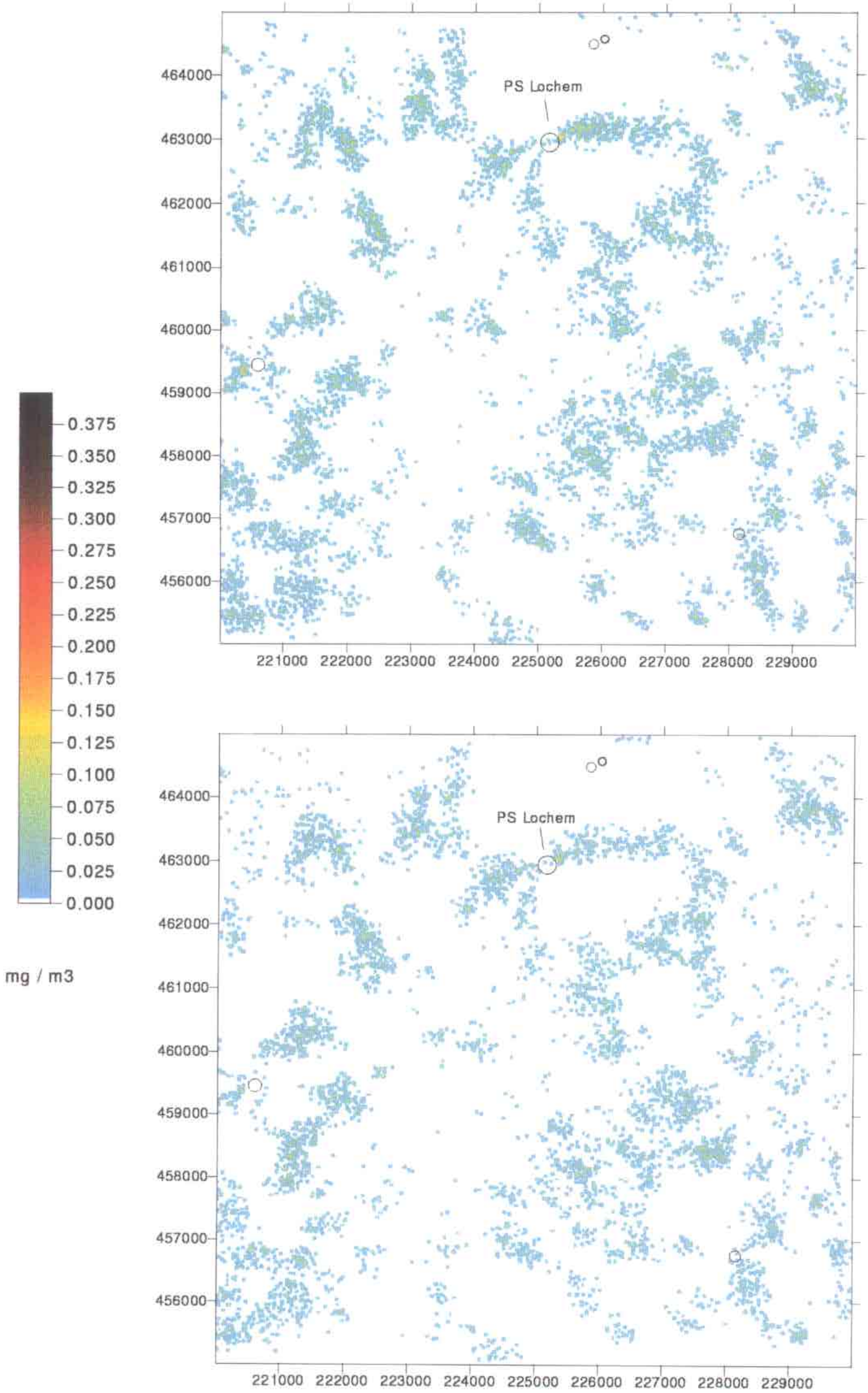


Figure 18 Concentration 20 m below groundwater level at 30 year (above) and 40 year (below)

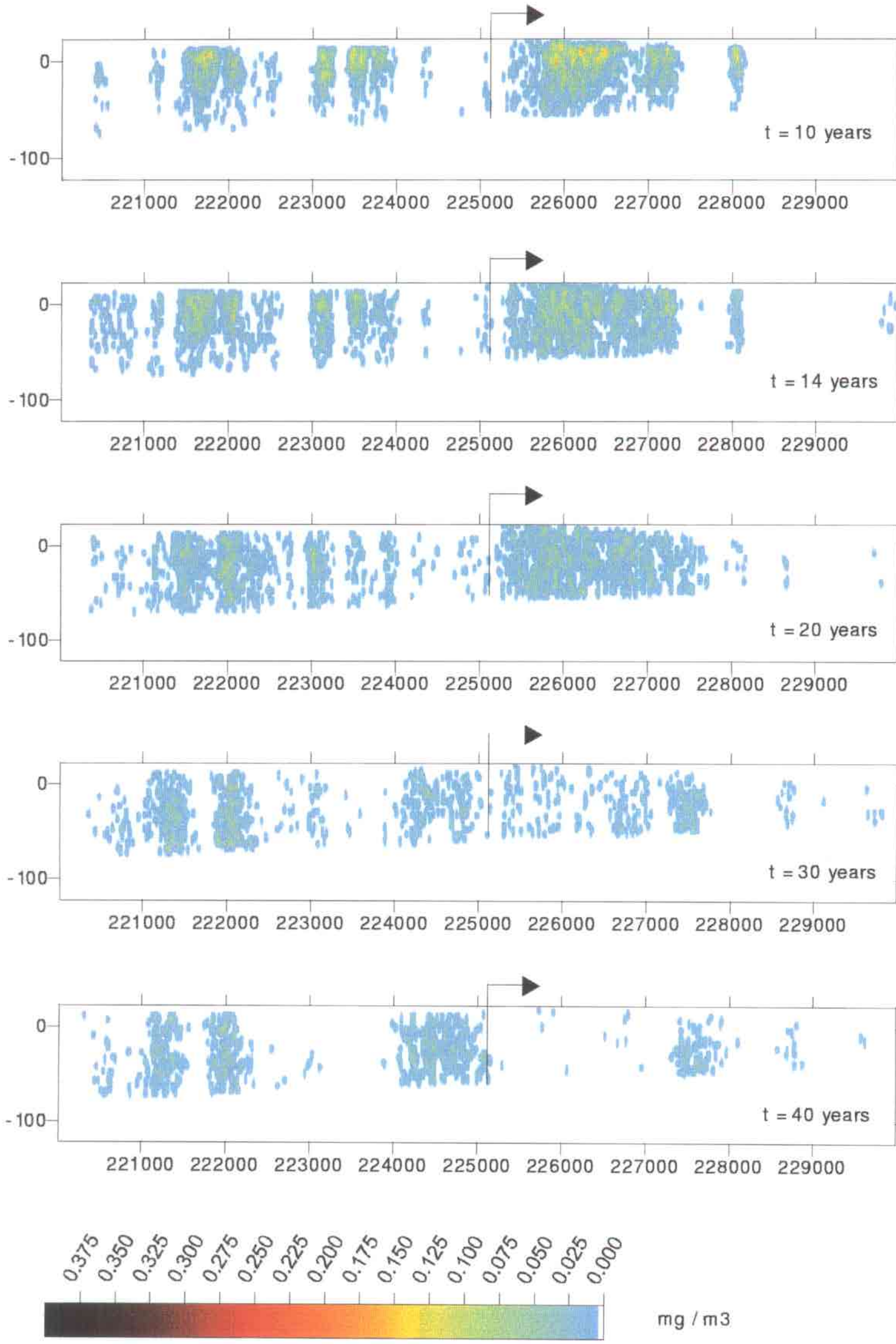


Figure 19 Concentration in cross-section B-B' (see fig 15) for several moments in time (scenario 1). Vertical axis denotes depth in m with respect to NAP.

5.4 Scenario 2

In this scenario a continuous use of the pesticide is assumed. Obviously, the gradual decrease in concentration, as seen in the previous scenario, is absent now. The spatial distribution of the concentration is shown in the figures 20, 21 and 22 for a depth of 10 meter below groundwater level and in the figures 23, 24 and 25 for a depth of 20 meter.

The breakthrough curve for PS Lochem is presented in figure 26. The curve ends at t = 60 years, since the simulation has been terminated here. The concentration in the well is expected to rise further for times larger than 60 years. Earlier investigations (Kovar et al., 1996) have shown that, for a uniform leaching rate in the capture area of the well at Lochem, the concentration reaches a stationary level after approximately 180 years. For the present problem it was not economical in terms of computer time to continue the simulation that long.

A vertical cross-section, located along the line *B-B'* (see figure 15), is given in figure 27. This figure gives the distribution of the concentration for times t equal to 10, 20, 30 and 40 years (figure 20). Note that the base of the aquifer is at a depth of 60-70 m -NAP and that the pesticides spread over the total aquifer thickness.

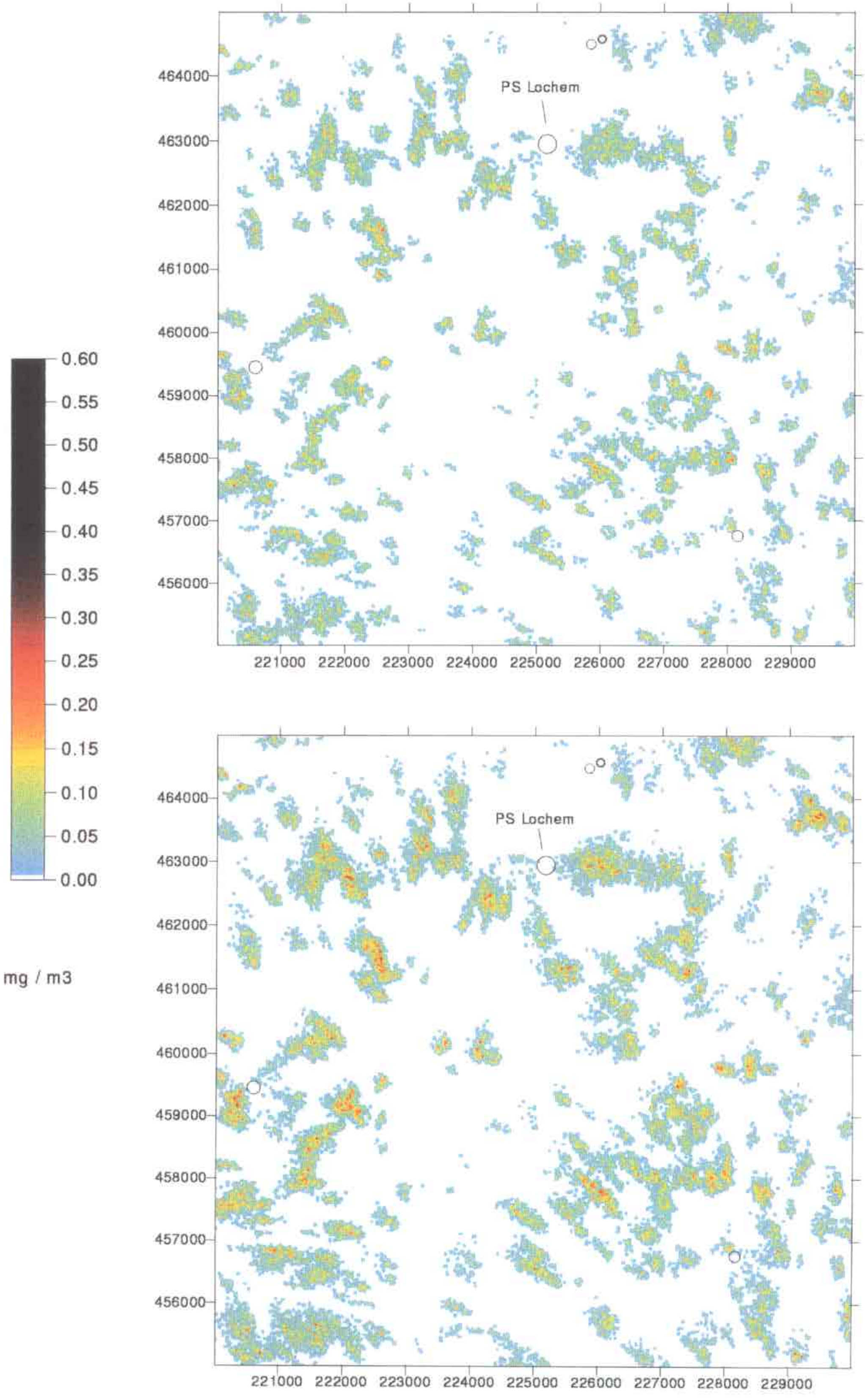


Figure 20 Concentration 10 m below groundwater level at 10 year (above) and 20 year (below) Scenario 2

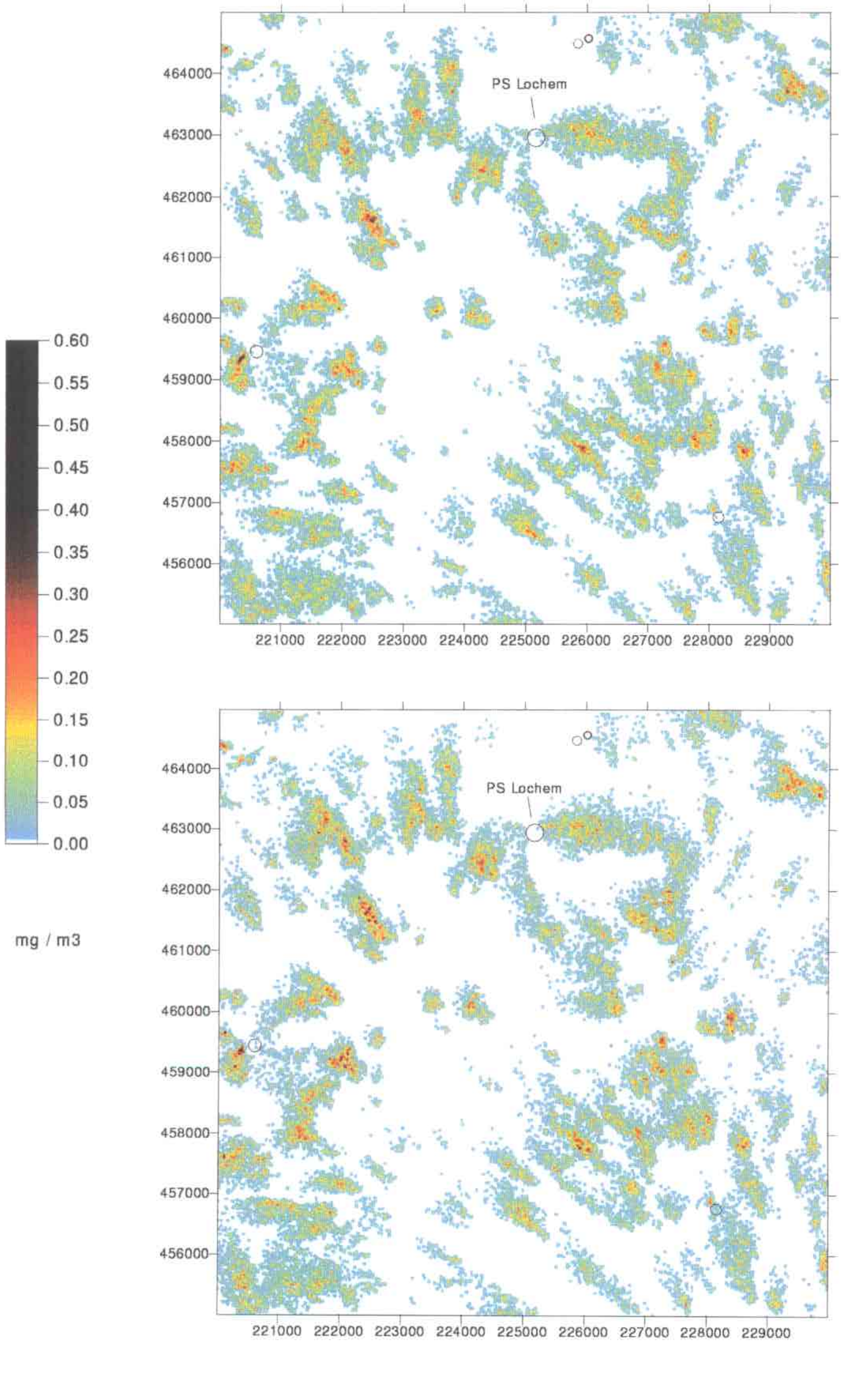


Figure 21 Concentration 10 m below groundwater level at 30 year (above) and 40 year (below) Scenario 2

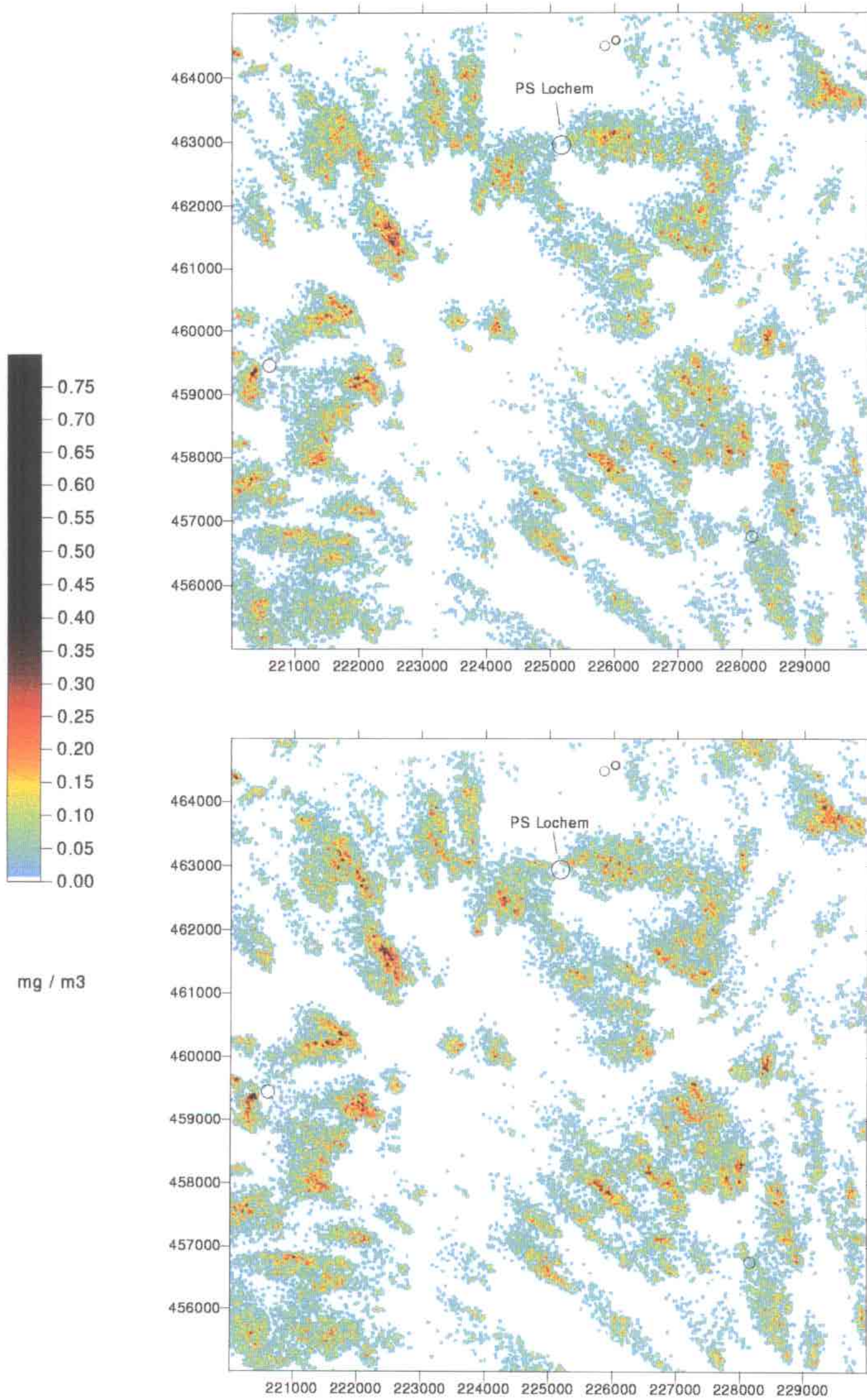


Figure 22 Concentration 10 m below groundwater level at 50 year (above) and 60 year (below) Scenario 2

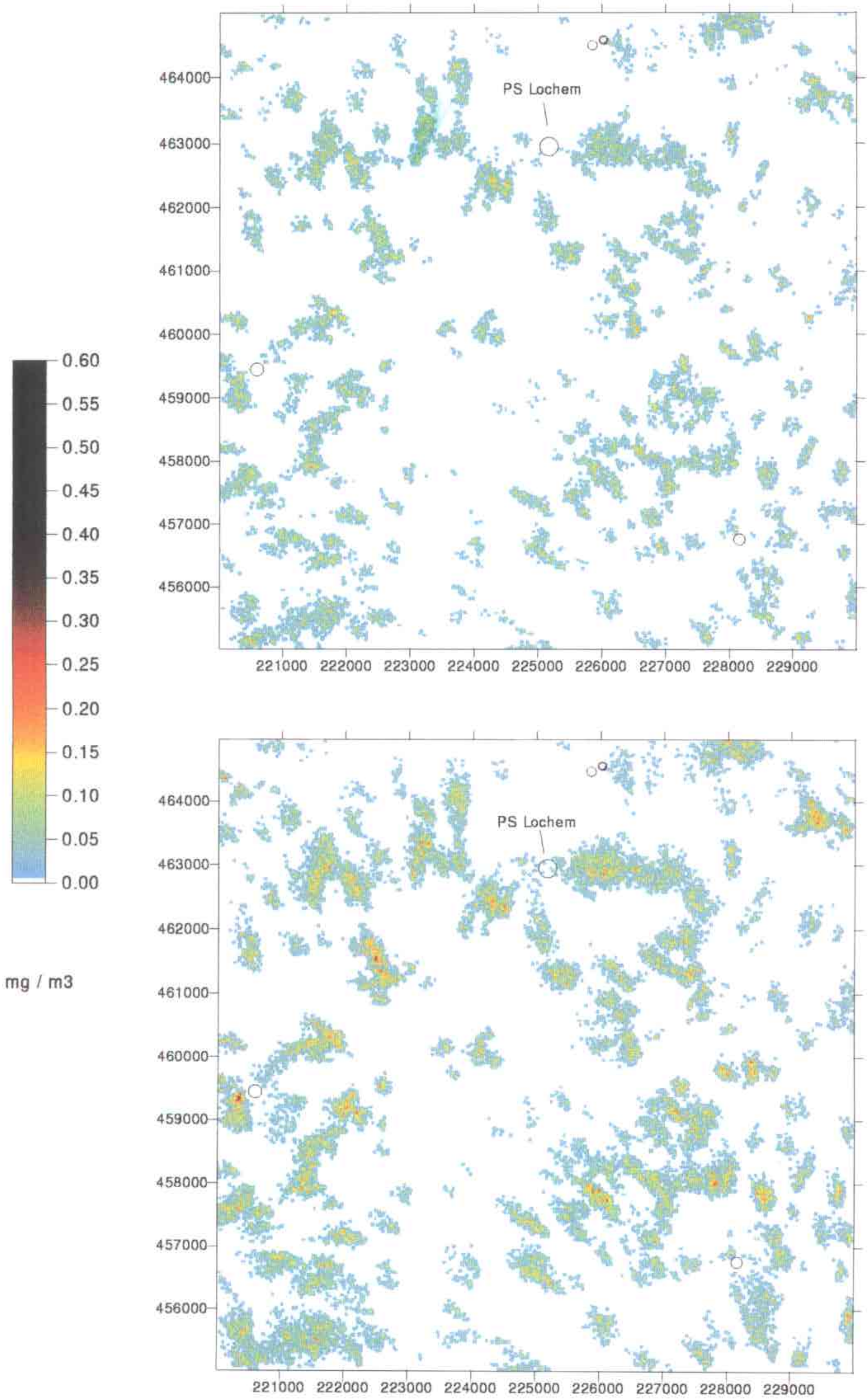


Figure 23 Concentration 20 m below groundwater level at 10 year (above) and 20 year (below) Scenario 2

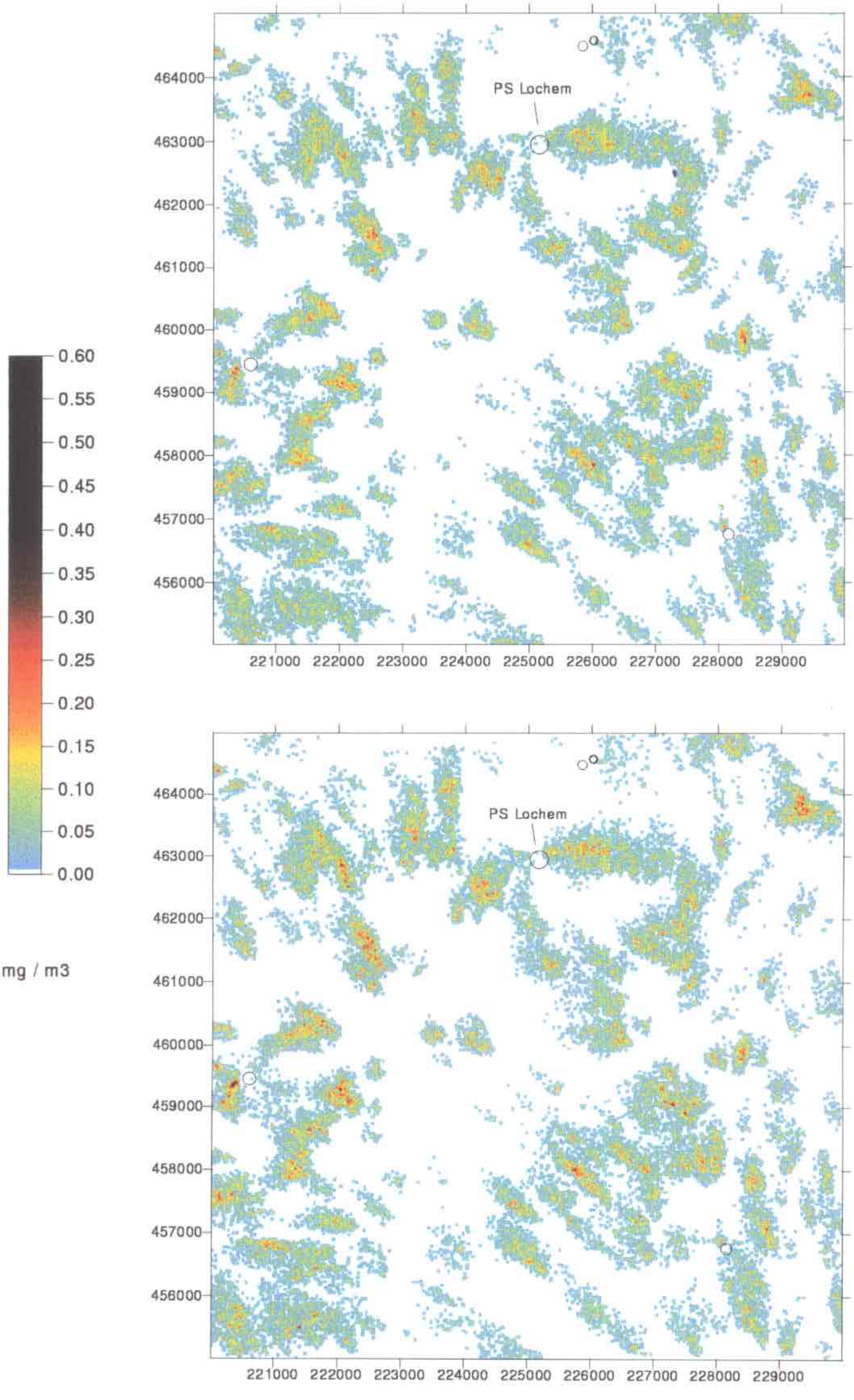


Figure 24 Concentration 20 m below groundwater level at 30 year (above) and 40 year (below) Scenario 2

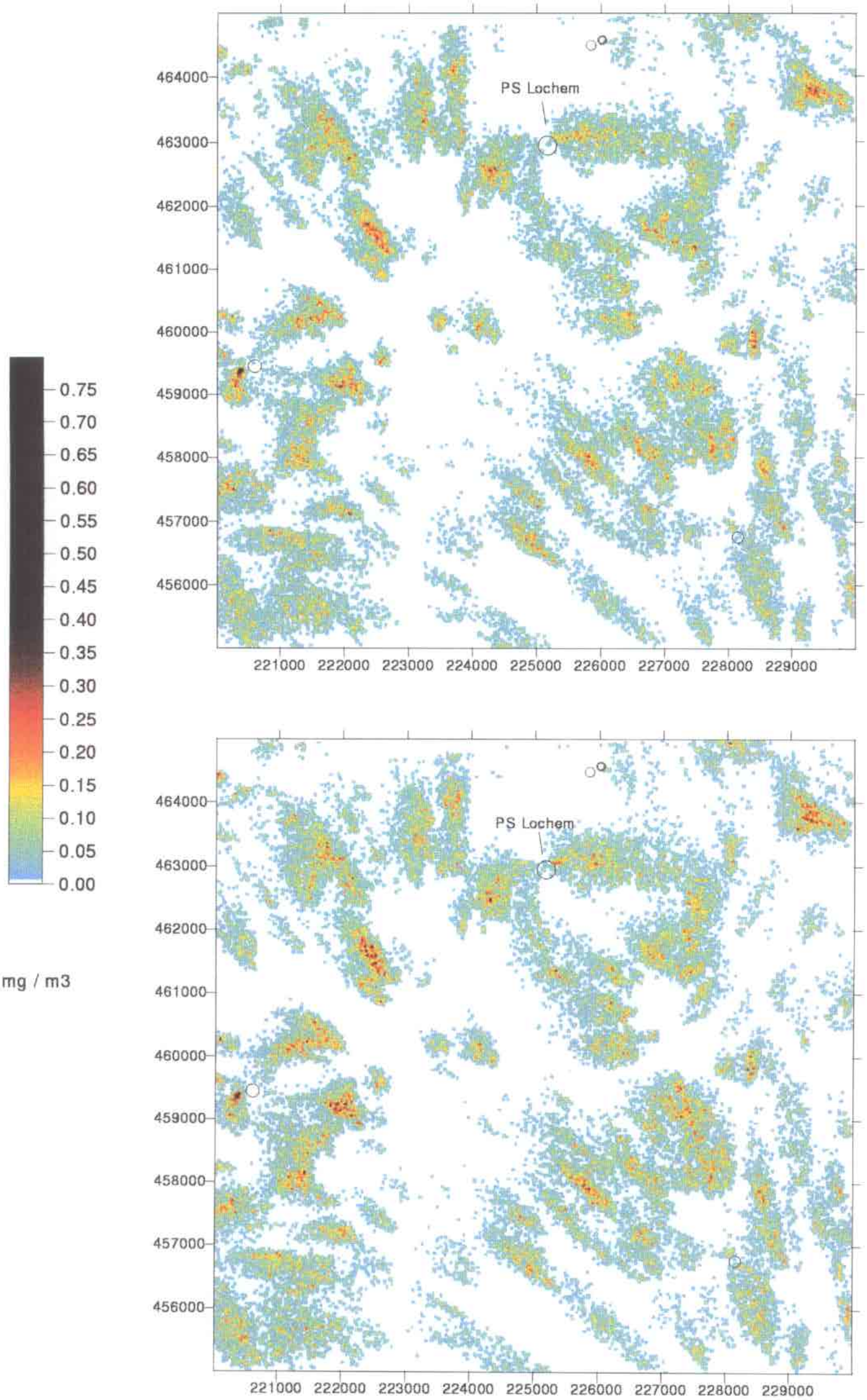


Figure 25 Concentration 20 m below groundwater level at 50 year (above) and 60 year (below) Scenario 2

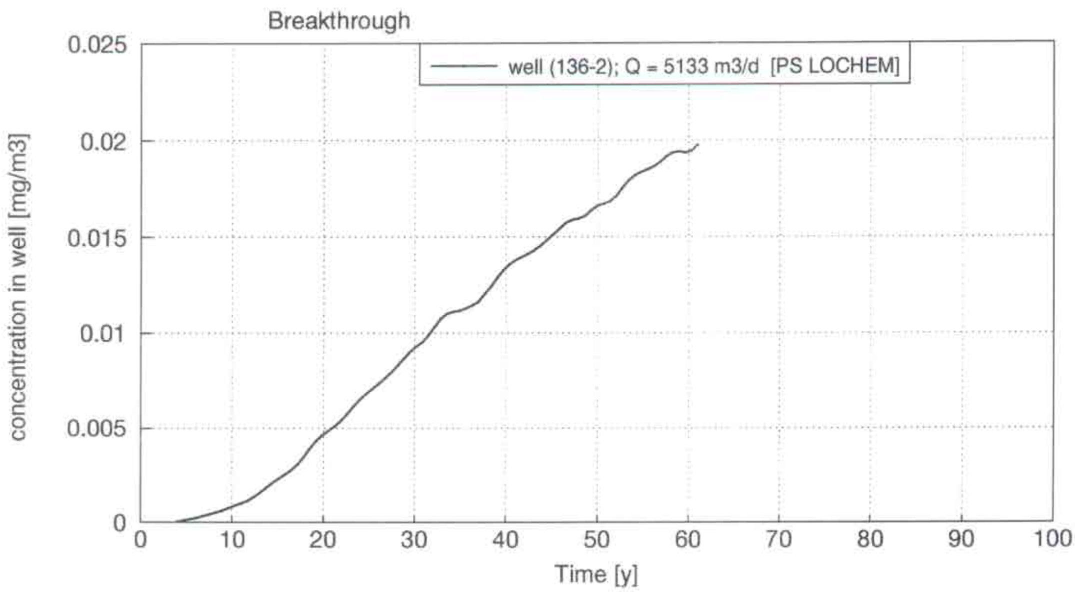


Figure 26 Breakthrough curve at PS Lochem for scenario 2

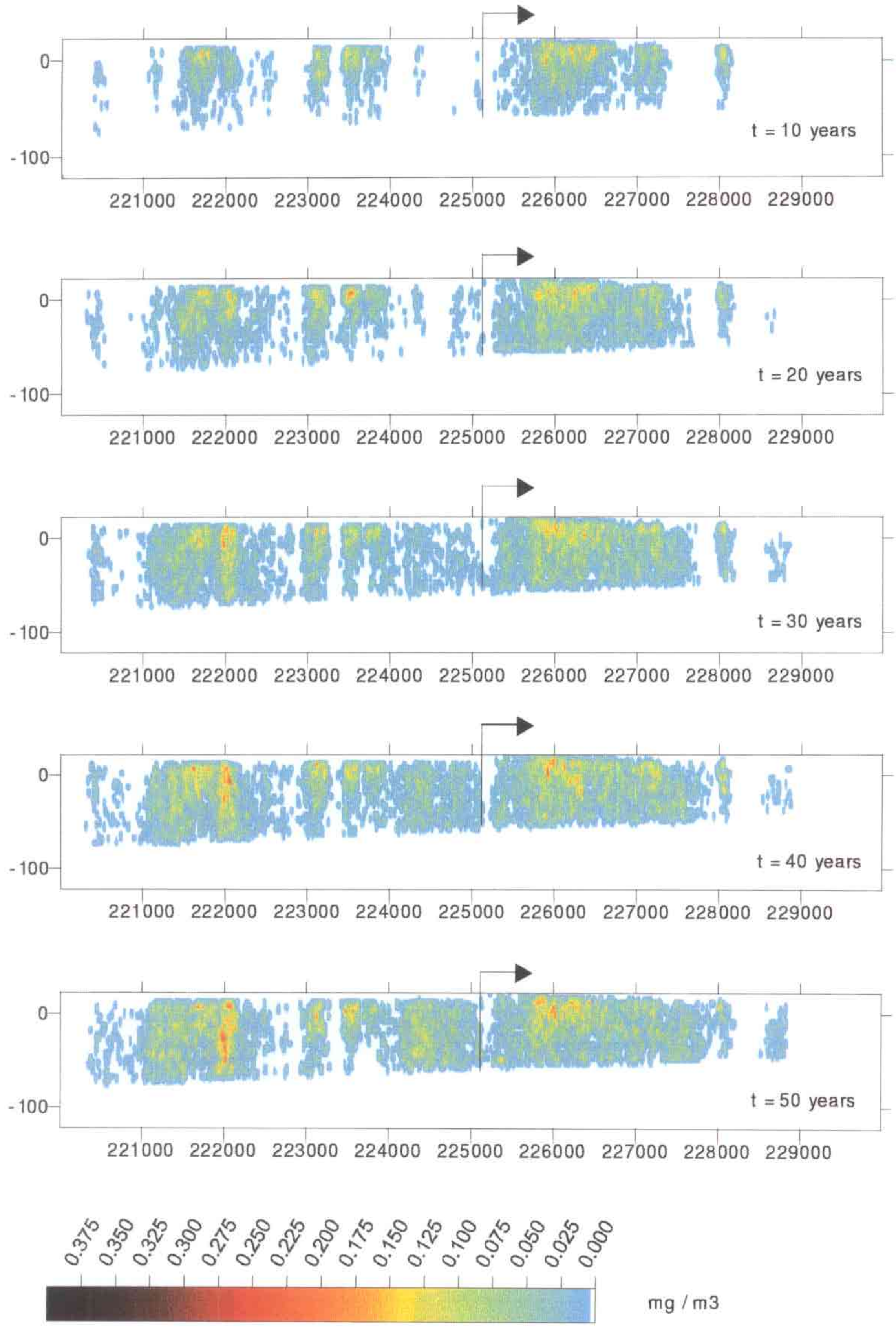


Figure 27 Concentration in cross-section B-B' (see fig 15, for several moments in time (scenario 2) Vertical axis denotes depth in m with respect to NAP.

5.5 *Scenario 3*

In scenario 3 attention is focused on the effect of crop rotation. The pesticide is applied during one year in a series of 4 years. This pattern is maintained during the total simulation period. The average load of pesticides to the groundwater is 1/4 of the load in scenario 2. If the dilution in the groundwater is optimal, the concentrations in this scenario will be 1/4 of the concentrations in scenario 2. However, the leaching of pesticides is inhomogeneous in time. A point of interest here is, to what extent fluctuations in the concentration of the infiltrating water are still present in the groundwater.

The spatial distribution at a depth of 10 m and 20 m below groundwater level is displayed in the figures 28 - 33, for various moments in time. A cross-section with the concentration distribution for various moments in time is given in figure 34.

To study the variation in time, two monitoring points have been introduced, for which breakthrough curves have been calculated (figure 35-38). Also profiles of the concentration versus depth for various moments in time (figures 39 and 40) have been calculated. These calculations have been repeated for purely advective transport, in order to demonstrate the effect of dispersion more clearly. The coordinates of monitoring point-1 are x = 226000 m, y = 463000 m and z = 2.5 m. The vertical position of the monitoring point corresponds with a depth of 10 m below groundwater level. Monitoring point-2 is at the same horizontal position, but at a depth of 20 m below the groundwater table. The horizontal position is indicated in figure 28. The monitoring point is not a point in the mathematical sense, but represents a cell with horizontal dimensions 25 x 25 m^2 and a vertical height of 5 meter. Figure 36 gives the breakthrough in case dispersion is absent (advective transport), while the figures 35 gives the breakthrough for advective-dispersive transport. The periodic variation in the leaching pattern (period of 1460 days) can still be recognized in the breakthrough curves. The curves in figures 35 and 37 show what happens if dispersion is taken into account. Due to transverse and longitudinal dispersion the pesticides are spread out over a volume of groundwater that is larger than in the case of advection only. This leads to a reduction of the average concentration level. Moreover, longitudinal dispersion causes a fraction of the contaminants to travel faster along the streamlines, than the advective travel time. This results in a reduction of the first breakthrough times.

Another illustration of the influence of dispersion is given in figures 39, 40 and 41. Figures 39 and 40 present vertical concentration profiles for various moments in time. In the case of pure advection (fig 39) the pesticides do not occur at depths below 20 m -NAP, but when dispersion is included (fig 40), the pesticides are found over the total height of the aquifer . This situation can be explained at best with a sketch of the flow pattern (fig 41). Below 20 m -NAP the origin of the groundwater is either from non-maize fields, or from maize fields at a distance too far away to be covered by advective transport (fig 41a). The presence of pesticides below the 20 m -NAP in the advective-dispersive case (fig 41b) is due to transverse dispersion of the pesticide-plume infiltrating at near-by maize fields or due to longitudinal dispersion of plumes from fields further away.

The breakthrough curve at the pumping well (PS-Lochem) is given in figure 42. Here, variations in time are hardly visible. At the well water enters with a wide range of residence times, or one may say the water particles have different 'ages' and therefore represent each period of the leaching pattern.

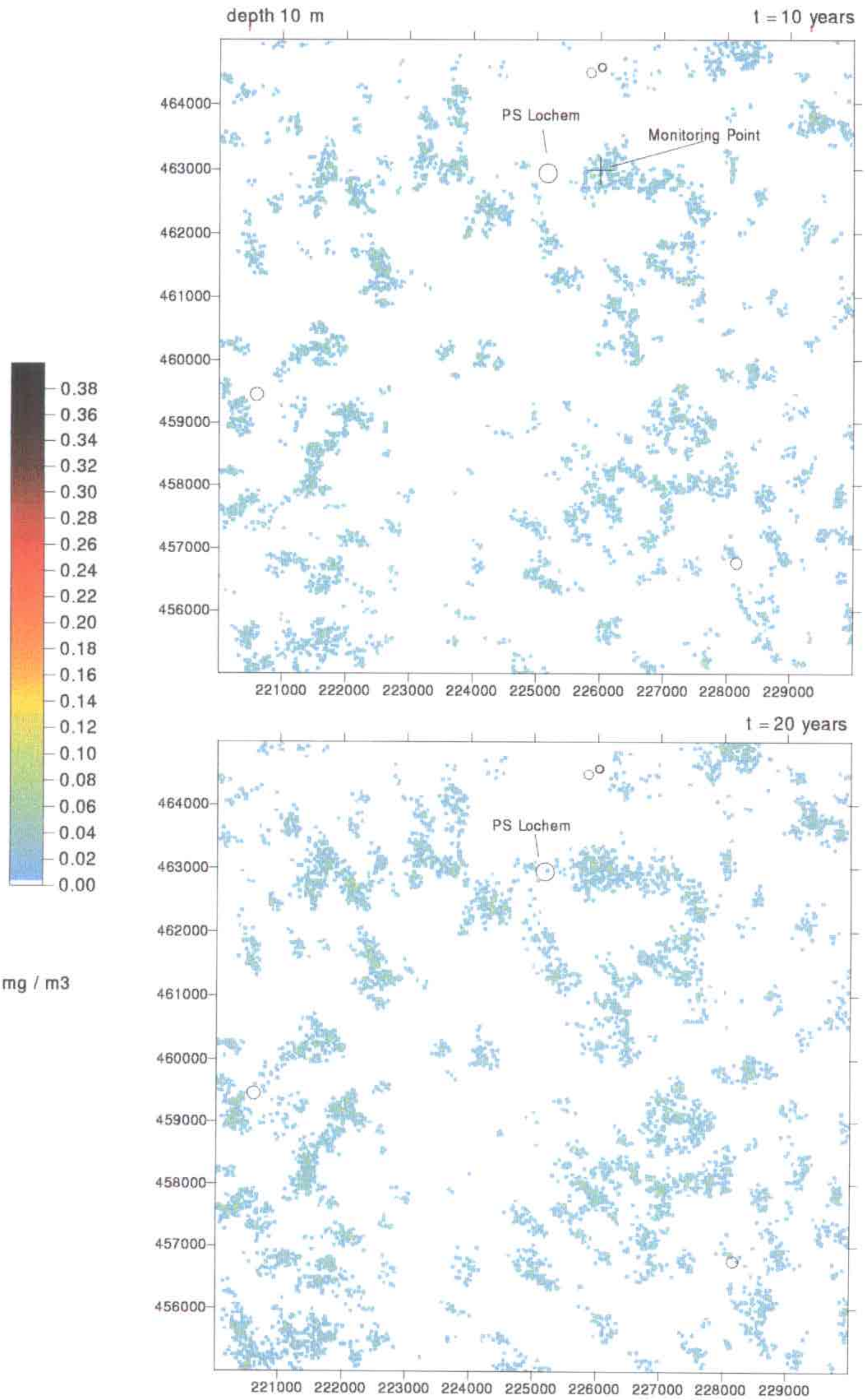


Figure 28 Concentration 10 m below groundwater level at 10 year (above) and 20 year (below); scenario 3

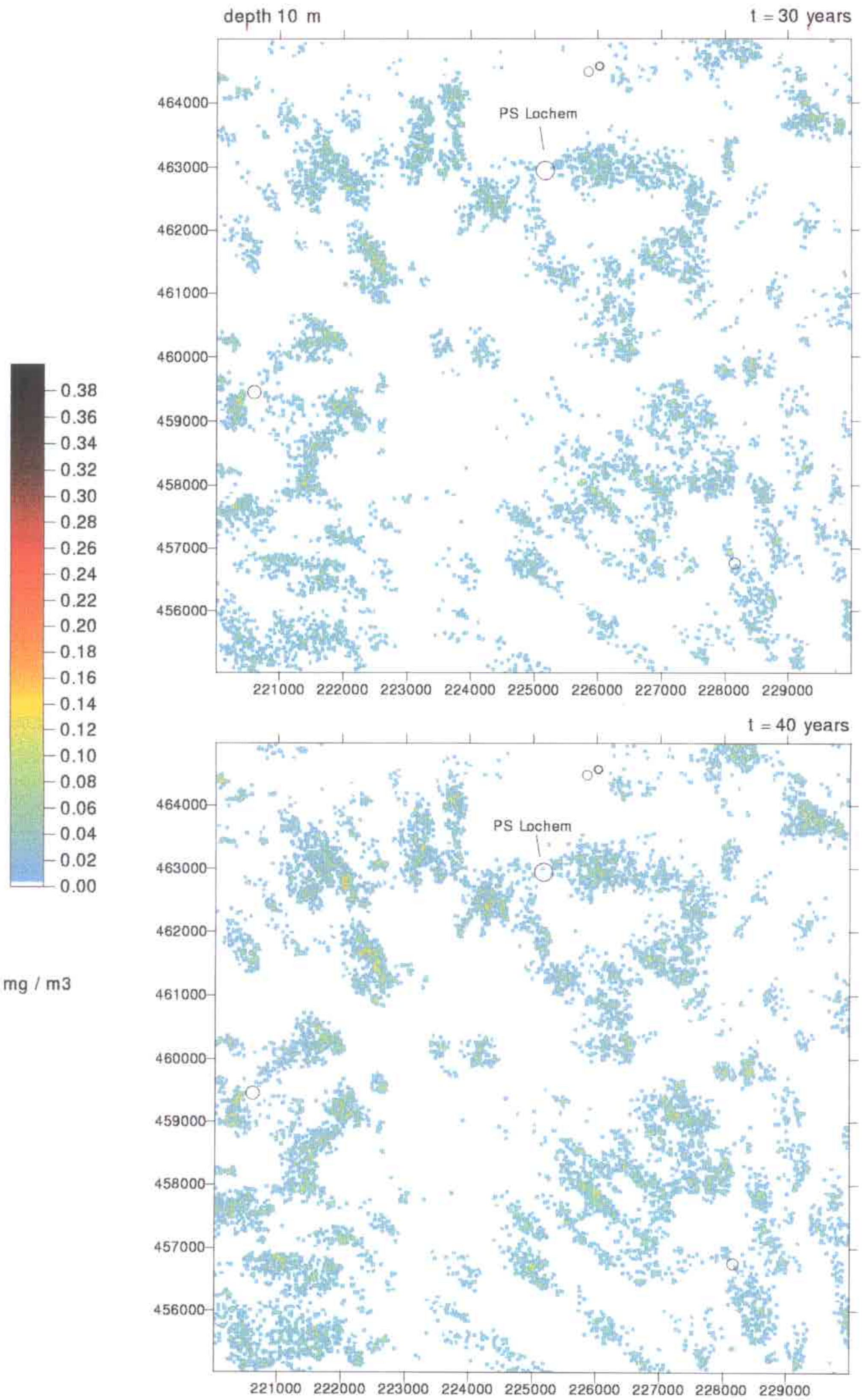


Figure 29 Concentration 10 m below groundwater level at 30 year (above) and 40 year (below); scenario 3

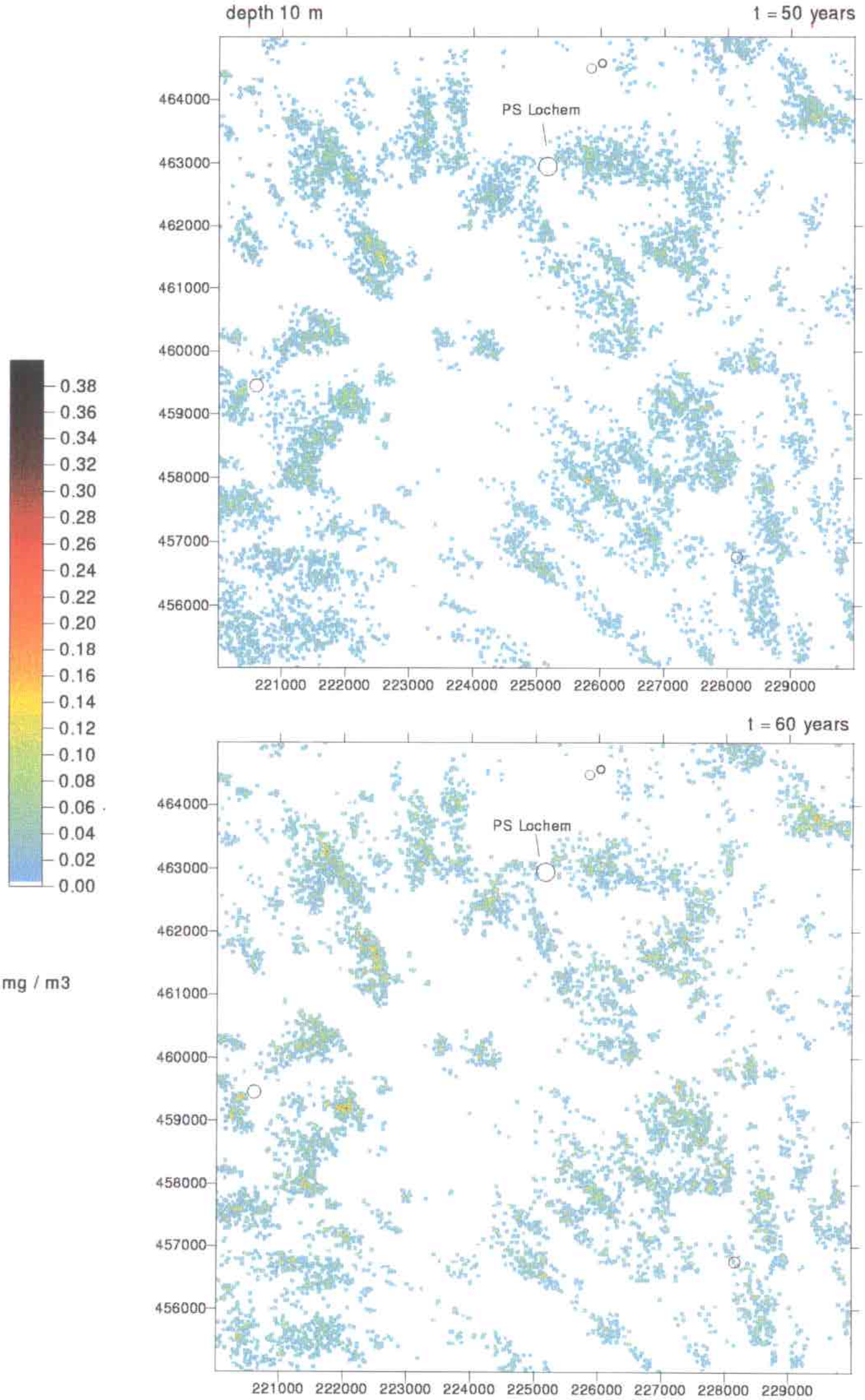


Figure 30 Concentration 10 m below groundwater level at 50 year (above) and 60 year (below); scenario 3

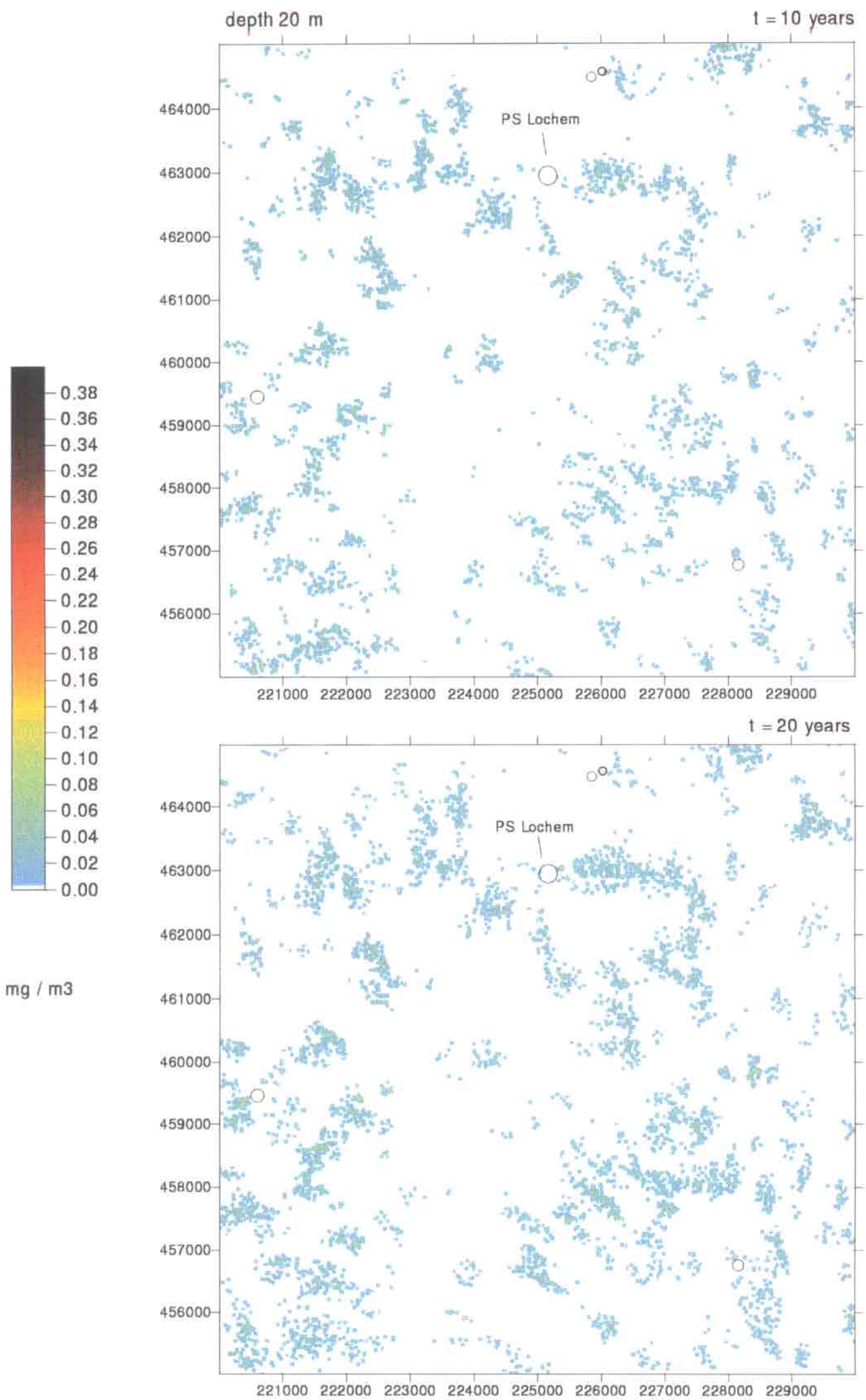


Figure 31 Concentration 20 m below groundwater level at 10 year (above) and 20 year (below); scenario 3

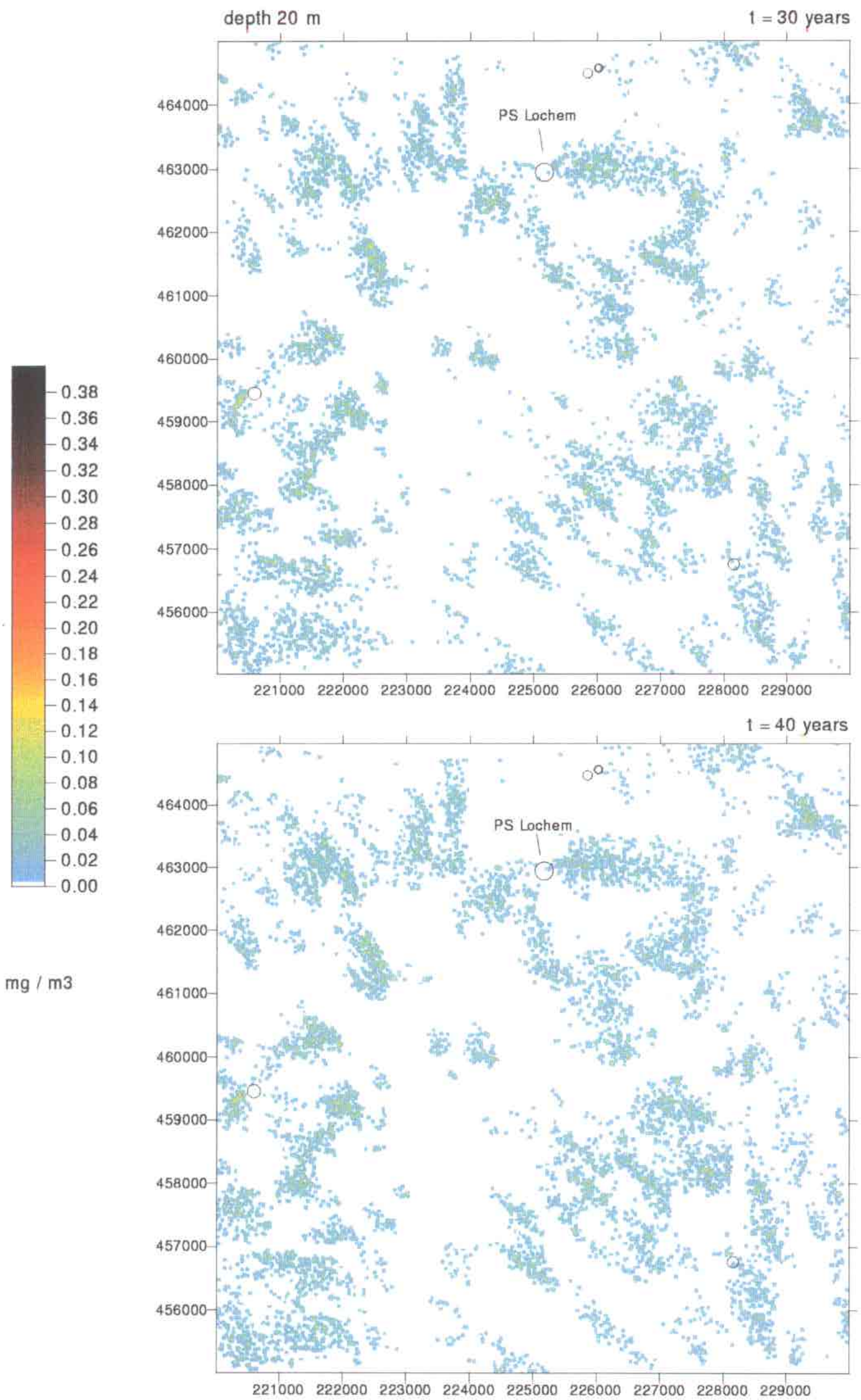


Figure 32 Concentration 20 m below groundwater level at 30 year (above) and 40 year (below); scenario 3

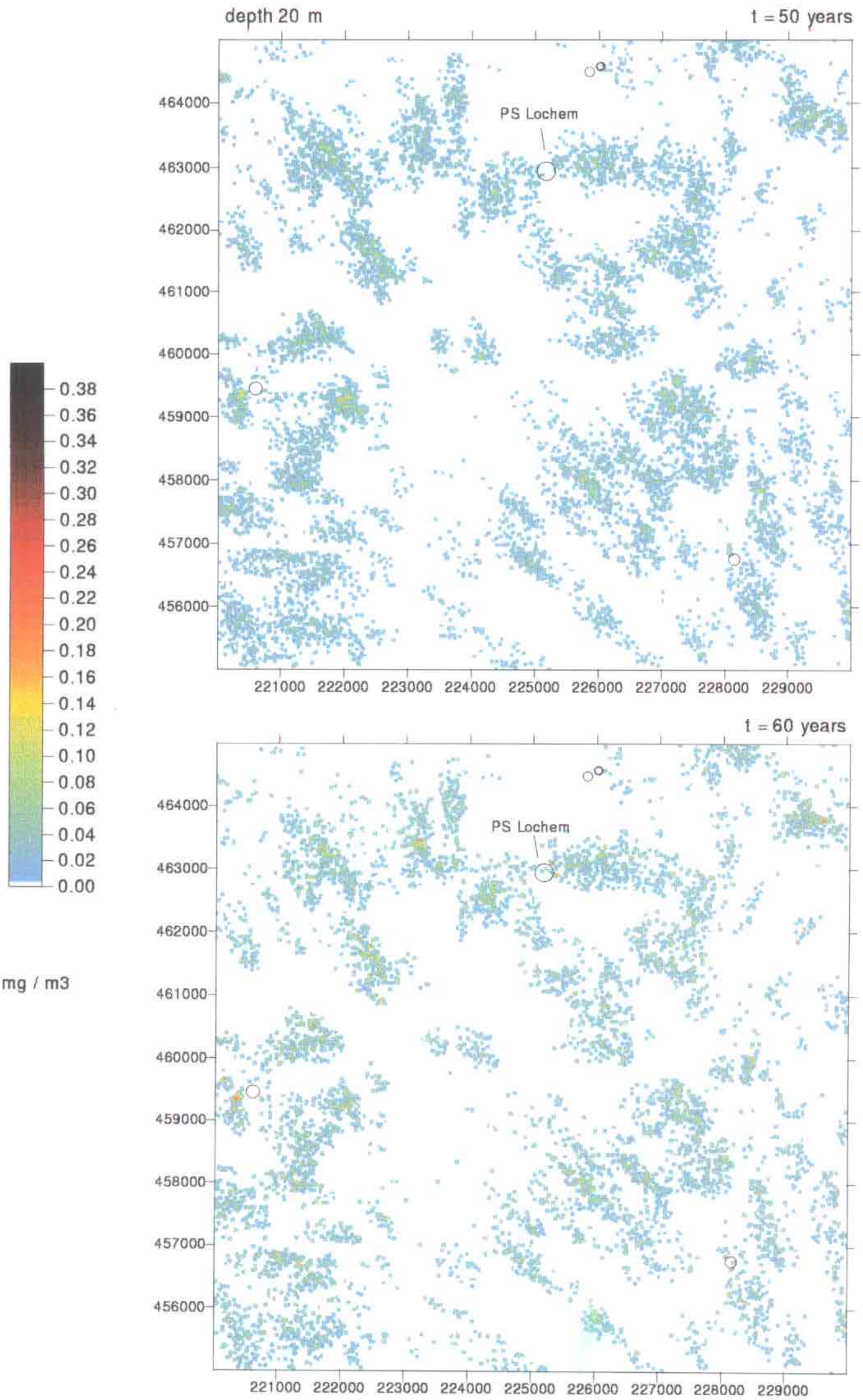


Figure 33 Concentration 20 m below groundwater level at 50 year (above) and 60 year (below); scenario 3

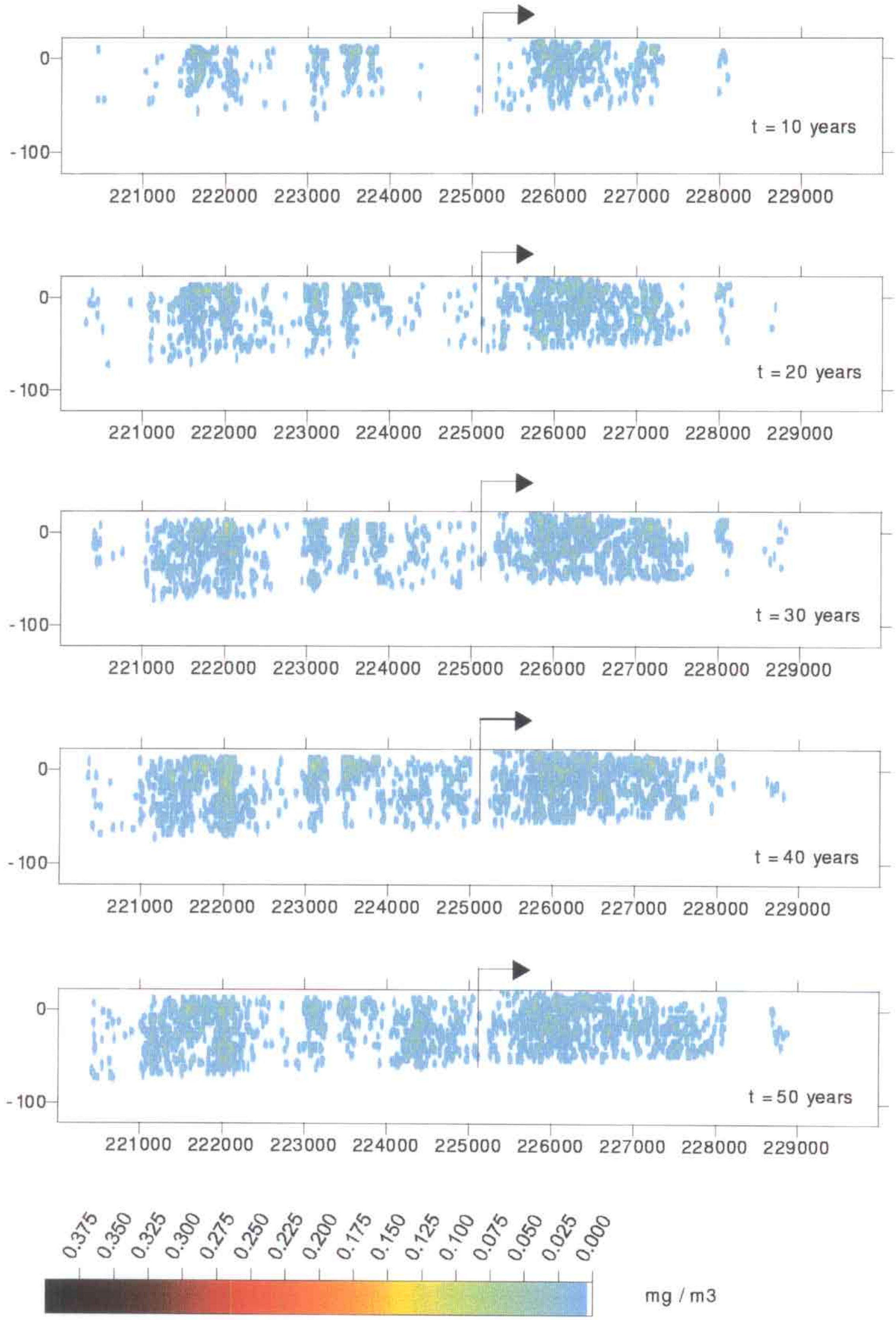


Figure 34 Concentration in cross-section B-B' for several moments in time (scenario 3)

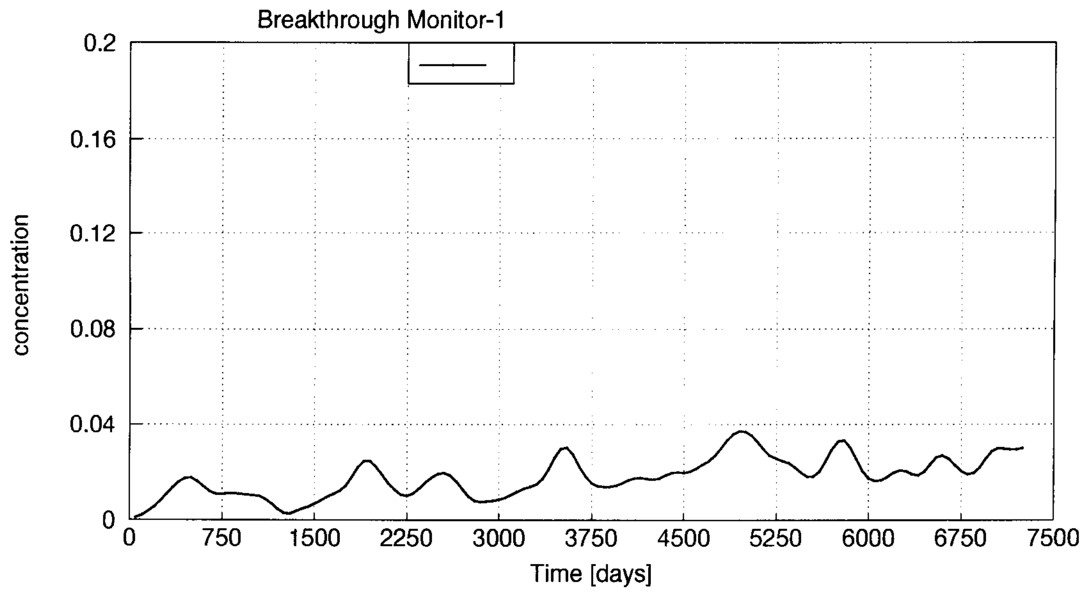


Figure 35 Breakthrough curve at Monitoring Point 1, 10 meter below groundwater. (scenario 3) Advective and dispersive transport.

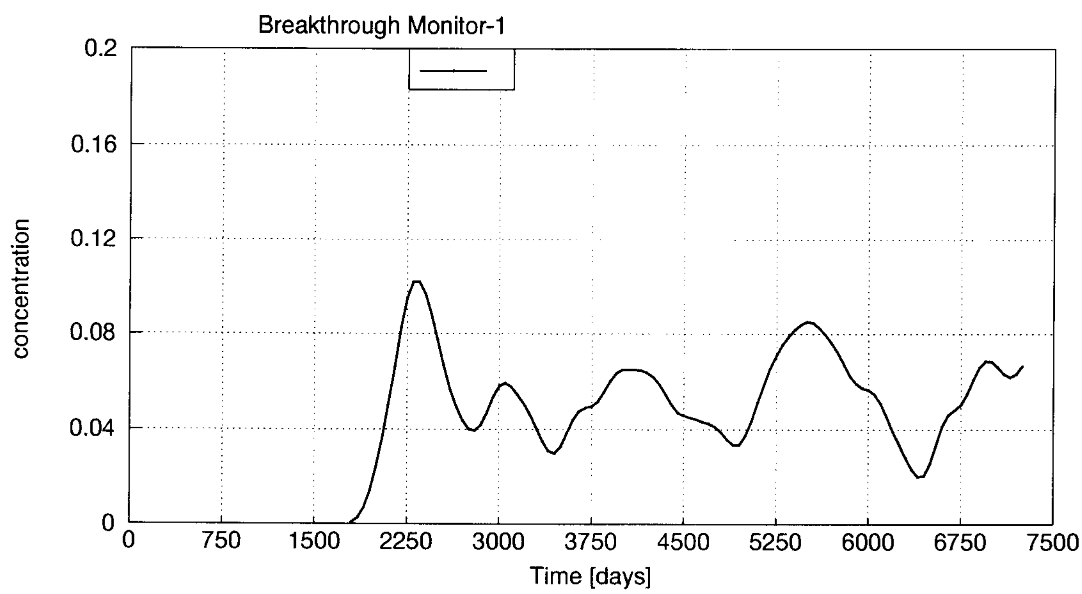


Figure 36 Breakthrough curve at Monitoring Point 1, 10 meter below groundwater. (scenario 3). Advective transport only

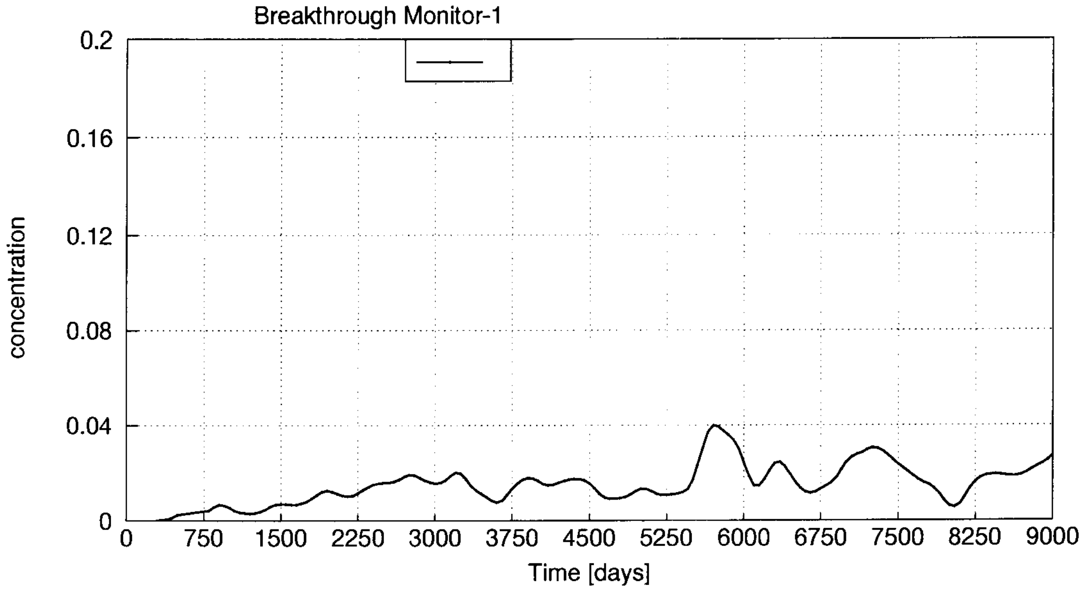


Figure 37 Breakthrough curve at Monitoring Point 1, 20 meter below groundwater. (scenario 3) Advective and dispersive transport.

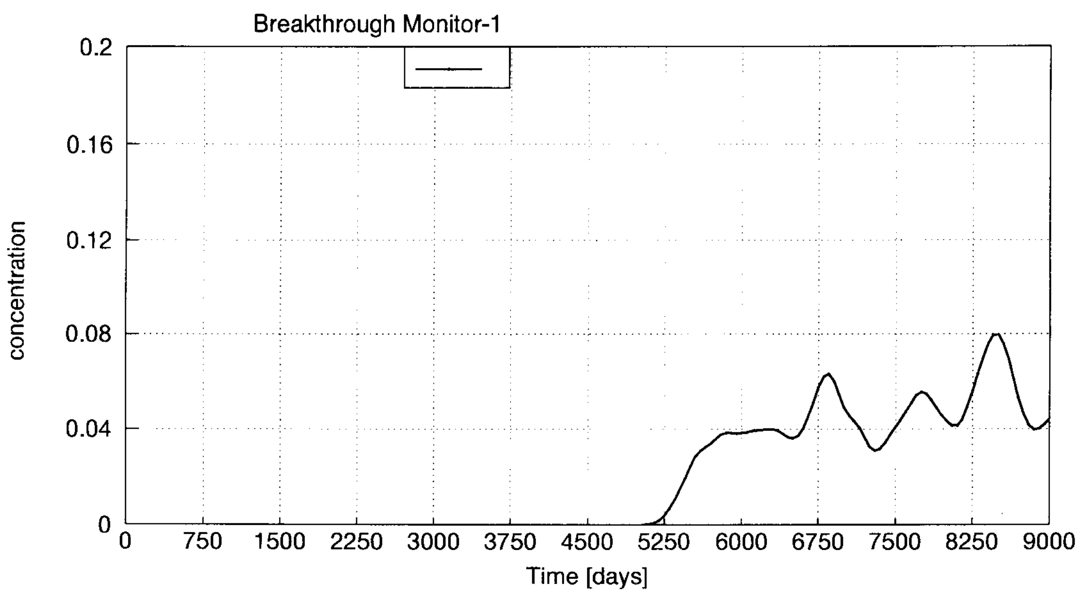


Figure 38 Breakthrough curve at Monitoring Point 1, 20 meter below groundwater. (scenario 3) Advective transport only.

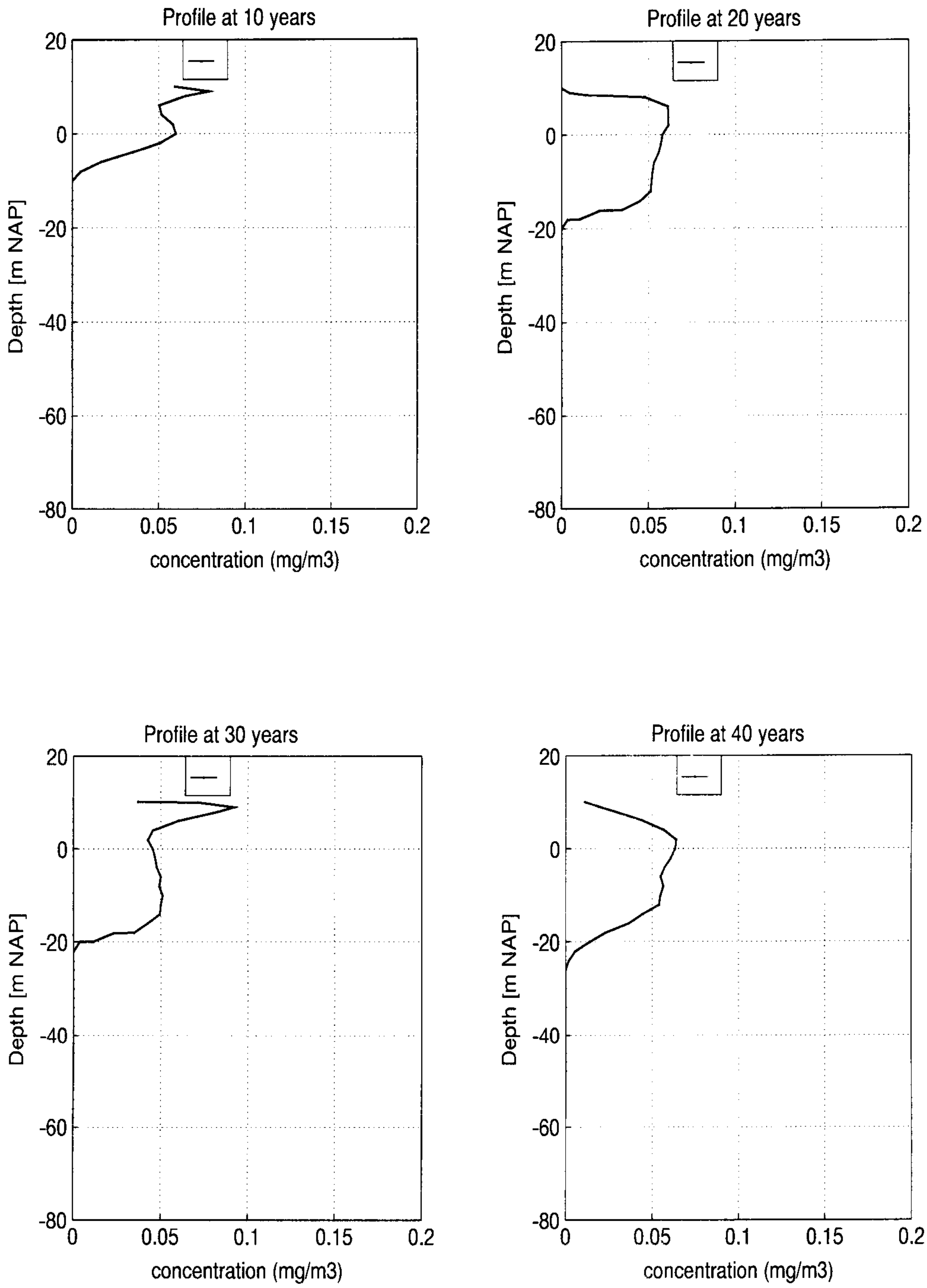


Figure 39 Concentration versus depth for various moments in time, considering advection only

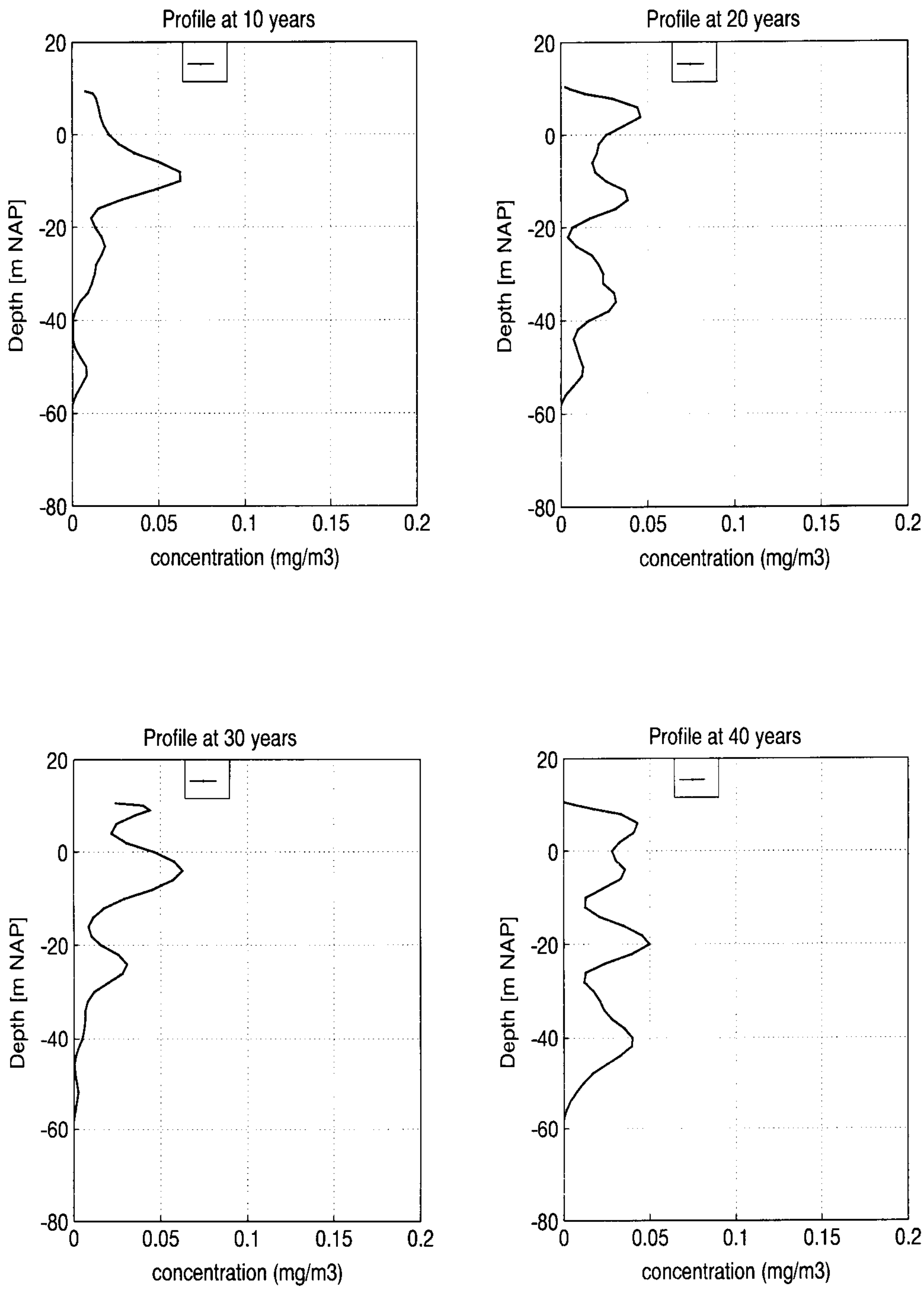
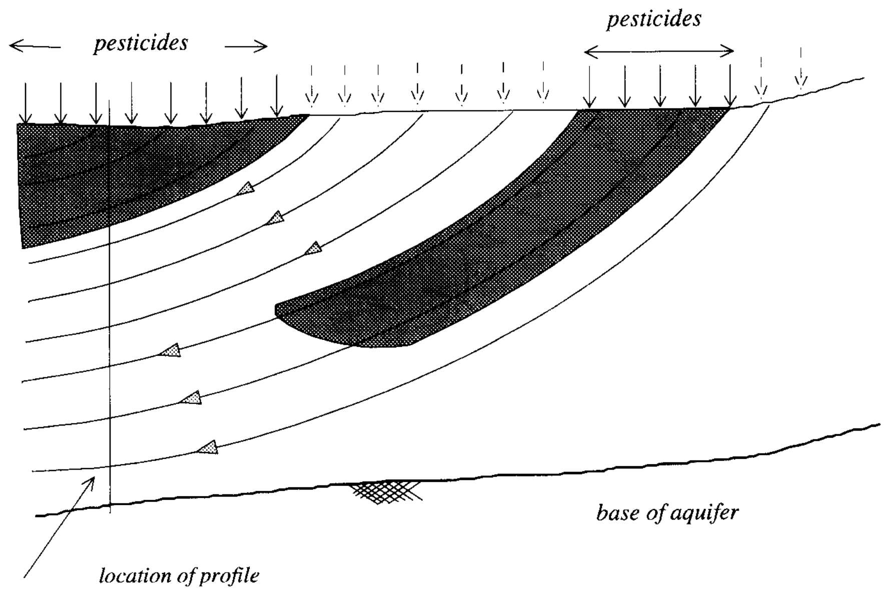
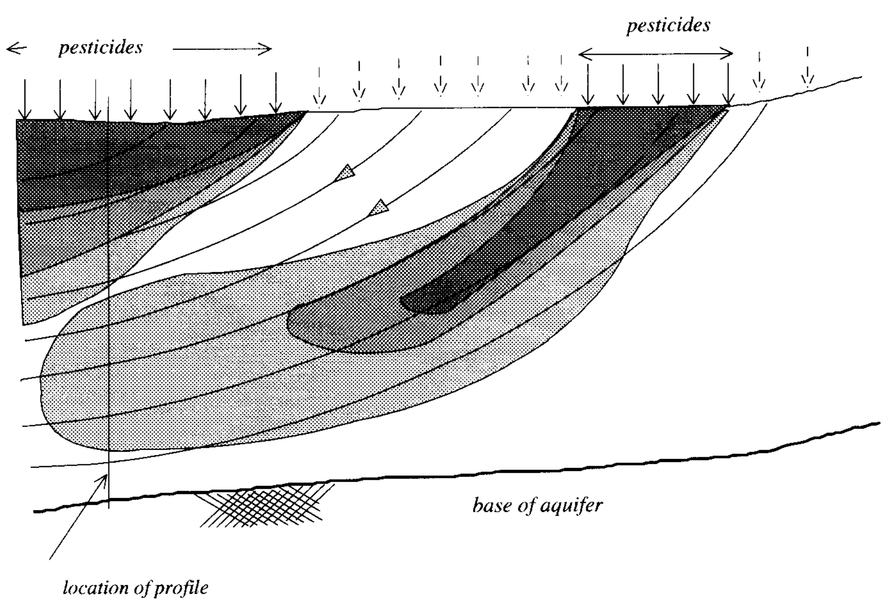


Figure 40 Concentration versus depth for various moments in time, considering advection-dispersion.



(a)



(b)

Figure 41 Sketch of pesticide-plumes for advection (a) and advection-dispersion (b)

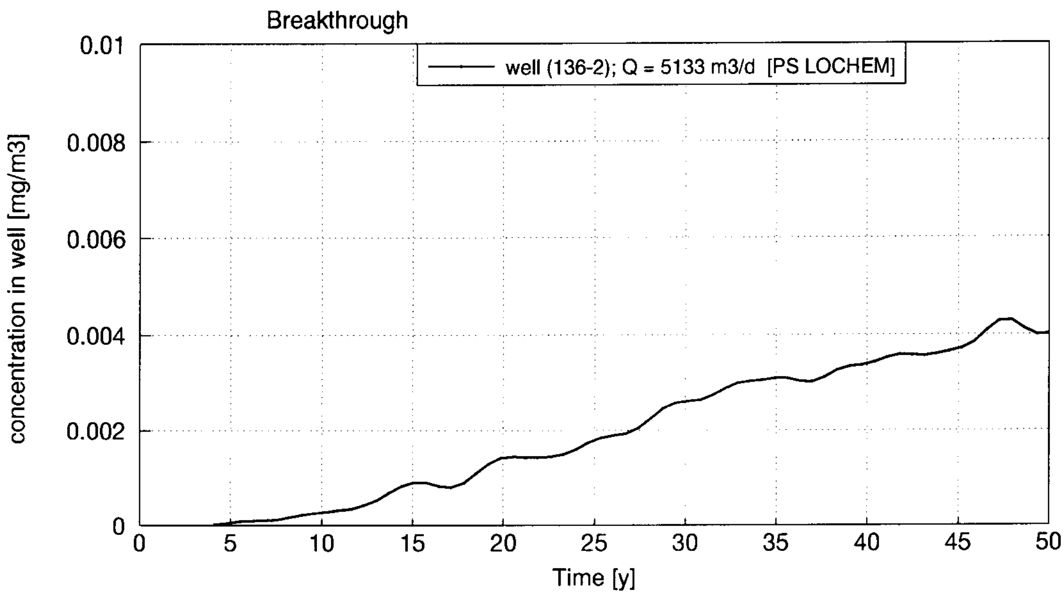


Figure 42 Breakthrough curve at Well of Pumping Station Lochem (scenario 3) Advective dispersion transport .

6 DISCUSSION AND CONCLUSIONS

This report addresses the question whether dispersion contributes substantially in reducing concentrations of pesticides in the saturated zone. It was decided to investigate this question by scenario type computer simulations for a rather well known area in the Netherlands. Well known here means that the hydrological situation has been investigated in great detail. The area around pumping station Lochem fulfills this criterion. Also, it has the advantage that both recharge and seepage situations occur and, therefore, effects of seepage and recharge on the possible dilution can be included in the study. The land use around pumping station Lochem is a scattering of arable land (mainly maize) and grassland, which allows us to study the effects of spatial variation in pesticide loads in a realistic way.

The simulations are carried out for a hypothetical, but realistic load at the phreatic surface below arable land. It has been assumed that no pesticides are used on grassland, and that on maize-land 0.1% of the amount applied on the fields reaches the groundwater. Given the fixed load of 0.1 mg/m^2, the concentrations will depend on the amount of recharge water. In the uppermost groundwater in the study area simulated concentrations are up to 0.8 $\mu g\ L^{-1}$. In several monitoring programs pesticide concentrations at this level have been observed, so the concentration range is realistic. For a better understanding of the following, it is noted that conservative behavior of the pesticide is assumed, i.e. bio-transformation and sorption are neglected.

The simulations demonstrate that concentrations of pesticides decrease with increasing depth. The process responsible for the reduction is dispersion, since advection alone does not affect groundwater concentrations. The highest concentrations in the groundwater are expected for scenario 2, where pesticides are applied on the arable fields for a long period (60 years). At a depth of 10 m below the groundwater level the concentrations are lower than at the groundwater level. The reduction in concentration with depth is observed at best at the boundaries between grassland and arable land. The concentrations increase with time, as this scenario considers a continuous input of pesticides. At the end of the simulation period (60 years), concentrations are still below input concentrations, although at a few places the concentrations are close to the input concentration (around 0.75 $\mu g\ L^{-1}$). The final decrease is only a few per cent at these places. At 20 m below the groundwater level it takes more time to reach this level, but at the end of the simulation period also at this depth concentrations up to 0.75 $\mu g\ L^{-1}$ occur.

The normal duration of a registration period for pesticides is 10 years. If a compound is used for such a limited period (scenario 1), the concentrations will not reach the levels from scenario 2. At 10 m below the groundwater level simulated concentrations up to 35% of the input concentrations occur. With respect to the reduction in concentration, one must keep in mind that, although dispersion reduces concentrations, it does not reduce the total amount of pesticides in the aquifer. The reduction of concentration by dispersion occurs at the expense of a larger contaminated area, while it also results in a longer time for a recovery of the groundwater quality in that area. The simulations indicate after a limited registration period of 10 years, it may take several decades to obtain groundwater that is free of pesticides. One may conclude that a registration period of 10 years is sufficiently long to contaminate large areas, while the effect of dilution, though significant in some parts, may be limited in other parts of the area.

The concentrations in the abstraction well (Pumping station Lochem) show a comparatively large dilution. In scenario 2 at the end of the simulation period the pesticide concentration in the pumping well is almost 0.02 $\mu g\ L^{-1}$, but still increases. The average concentration of the groundwater recharge in the capture zone of the well is approximately 0.25 $\mu g\ L^{-1}$ (calculated concentration). Given the concentration in the recharge and the land use in the capture zone, it is not expected that the steady state concentration will become higher than 0.1 $\mu g\ L^{-1}$. The expected dilution is at least a factor of 2.5. However, the dilution here is due to the fact that in the well both contaminated and uncontaminated groundwater come together. The role of dispersion is not essential. The concentration of the water in the pumping well depends, apart from the concentrations in the aquifer, on the volume ratio of contaminated and uncontaminated water. The effect of dispersion is that, on the one hand the amount of contaminated water increases, while on the other hand the concentration of contaminated water decreases. These effects compensate each other considerably.

Dispersive mixing also affects periodic fluctuations. When pesticides are applied during one year, followed by three pesticide-free years, fluctuations in the concentration may occur. To study the effect on periodic fluctuation a crop rotation scheme is devised (scenario 3). For such a scheme the total load of pesticides to the groundwater is reduced to $1/4^{th}$ of the level in the other scenarios, since the load of pesticides takes place only once in each rotation period. Calculations for an individual observation point (Monitoring point 1) show that, when dispersion is included, concentrations are much lower than when only advective transport is considered. Dispersion roughly diminishes the observed concentrations by a factor of 2. The fluctuations are considerably attenuated, but they are still noticeable. When the concentration maps of scenarios 2 and 3 are compared, it is obvious that concentrations in scenario 3 must be lower. However, a factor of 4, which corresponds to the difference in input, is not observed. At several spots, both at a depth of 10 and 20 m below the groundwater level, the attenuation is only a factor of 2, which agrees with the results of Monitoring point 1.

The conclusion is that in several parts of the area dispersion does not substantially lower the concentrations at a depth of 10 m below groundwater level. In some areas reductions of less than 5% are observed. If pesticide is applied in once every four years in a rotation scheme, the reduction in concentration may be as low as a factor of 2. In general, it can be stated that, especially for long term applications, dispersive mixing is no guarantee that concentrations at a depth of 10 m are reduced to below the critical level, when the concentration is above the critical level in the uppermost groundwater.

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