

On the scope and management of pesticide pollution of Swedish groundwater resources: The Scanian example

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Abstract Twenty-three south-Swedish public supply wells were studied to assess pesticide pollution of regional groundwater resources. Relations between pesticide occurrence, hydrogeology, and land use were analyzed using Kohonen's Self-Organizing Maps approach. Pesticides are demonstrated to be substantially present in regional groundwater, with detections in 18 wells. Concentrations above the drinking water threshold are confirmed for nine wells. Observations indicate considerable urban influence, and lagged effects of past, less restricted use. Modern, oxic waters from shallow, unconfined, unconsolidated or fracture-type bedrock aquifers appear particularly vulnerable. Least affected waters appear primarily associated with deeper wells, anoxic conditions, and more confined sediment aquifers lacking urban influence. Comprehensive, standardized monitoring of pesticides in groundwater need to be implemented nationwide to enable sound assessments of pollution status and trends, and to develop sound groundwater management plans in accordance with the Water Framework Directive. Further, existing water protection areas and associated regulations need to be reassessed.

Keywords Pesticides · Groundwater · Sweden · Water framework directive · Monitoring · Self-organizing maps

INTRODUCTION

Pesticides occur in groundwater worldwide. As groundwater tends to cycle slowly with limited potential for natural attenuation, and because pesticide use predictably will remain as global population and food demand increase, this occurrence is likely to persist well into the foreseeable future. It is a troublesome prospect, as pesticides can be toxic, and as groundwater supports ecosystems and food production worldwide, and accounts for about half of the world's potable water supply (WWAP 2009).

Upon use, pesticide transport into and throughout groundwater is essentially a matter of solute migration, transformation, and phase partitioning as governed by interactions between pesticide chemistry, soil biogeochemistry, and hydrogeological conditions dictating surface-groundwater connections, transport rates, and groundwater biogeochemistry (Barbash and Resek 1996). These are variable factors, as is pesticide use, and hence it is generally difficult to infer pollution status and extrapolate primary concerns from one setting to another. Regional studies of pesticide occurrence in groundwater are accordingly necessary to assess site-specific aquifer vulnerability and pollution risks, and to develop sound groundwater management plans. For EU member states, such assessments are mandatory under the Water Framework Directive (WFD; European Union 2000).

Comprehensive screening studies for pesticide occurrence in groundwater have been undertaken in many regions, e.g., the US (Gilliom 2007), Denmark (Thorling et al. 2012), New Zealand (Close and Skinner 2012), Norway (Haarstad and Ludvigsen 2007), and the Netherlands (Schipper et al. 2008). Between as well as within these studies, over time and space, measured pesticide concentrations can range orders of magnitudes, from the

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just detectable (currently ng L^{-1}) to several $\mu\text{g L}^{-1}$. Total detection frequencies (DFs) commonly range between 25 and 50 %, whereas DFs for concentrations at or above general environmental or health threshold criteria (e.g., the EC drinking water limit of $0.1 \mu\text{g L}^{-1}$) tend to be considerably lower, around 5–10 % (Schipper et al. 2008; Thorling et al. 2012). Differences in pollution extent are often noted between different types of wells, e.g., Haarstad and Ludvigsen (2007) showed considerably lower DFs for pesticides in public supply wells (PSWs), than in farm- and shallow-monitoring wells. Commonly detected substances include triazines such as atrazine and metabolites thereof (Gilliom 2007; Close and Skinner 2012; Thorling et al. 2012), and also, at least in Europe, 2,6-dichlorobenzamide (BAM; a metabolite of dichlobenil), aminomethylphosphonic acid (AMPA; metabolite of glyphosate), and bentazone (Schipper et al. 2008; Thorling et al. 2012). Both total and substance-specific DFs tend to be very dependent upon land-use, national pesticide regulations, and subsurface stratigraphy. In addition to agricultural pesticide use, urban weed control has also proven a much potent pollution contributor (Gilliom 2007; Malaguerra et al. 2012). Permeable and unconfined porous groundwater environments are typically found to be particularly vulnerable, however, for oxidizable pesticides, these types of environments have been found to provide better protection against leaching than semi-confining layers (Malaguerra et al. 2012). Local comparisons of pesticide pollution between completely different types of hydrogeological settings, i.e., sediment versus bedrock aquifers, are rare. Sampling depth, as a proxy of lag time between infiltration and observation, is commonly found to be inversely correlated to pesticide DF (e.g., Thorling et al. 2012). Relationships between estimates of this lag time through environmental tracer-based groundwater dating (Newman et al. 2010) and pesticide DF are generally more dubious, which is likely due to the fact that such ages, without detailed scrutiny, may be little representative of actual pesticide travel times, as that would require (i) negligible groundwater mixing and (ii) identical tracer and pesticide transport. Nevertheless, groundwater dating via e.g., tritium analysis (Solomon and Cook 2000) has proven useful for understanding general patterns of pesticide occurrence and absence (Kolpin et al. 1995), as it enables discrimination of predominantly modern groundwater which recharged after the onset of widespread pesticide use (post mid-1900 recharge), from predominantly old (pre mid-1900 recharge) groundwater.

In Sweden, about 50 % of drinking water is provided by groundwater (half artificially produced via surface water infiltration). The same figure is valid for the southernmost province of Sweden, Scania (Skåne), an intensively cultivated region for which groundwater resources have been

known to suffer from pesticide pollution for long (Maxe et al. 2003). Recently, results from a regional screening study ($n = 141$) initiated by the County Administrative Board of Scania proved pesticide occurrence in about half of the aquifers investigated, spurring questions of the potential trends and causal conditions, and highlighting a need for further investigations (Virgin 2012). However, to thoroughly assess the character and cause of pesticide pollution of Scanian, and effectively overall Swedish groundwater resources, is challenging due to a nationwide lack of comprehensive and consistent long-term groundwater monitoring data relevant to anthropogenic pollution concerns. This monitoring deficiency has previously been highlighted in relation to nutrient and pollutant loads in coastal areas (Destouni et al. 2008), with regards to heavy metal drainage from mining areas (Baresel and Destouni 2009), and more recently in view of general groundwater status and quality assessment in accordance with national environmental objectives and the WFD (European Commission 2012; SEPA 2013). Basically, the national groundwater monitoring network as set up and maintained by the national groundwater authority (SGU; the Geological Survey of Sweden), focuses on areas with minimal human impact to constrain background/reference conditions and does not involve pesticide analyses (Nordberg and Persson 1974; SGU 2013a). Quality control of groundwater used for drinking water is only mandated at the end-user, after potential treatment (SLVFS 2001:30). The only regular groundwater sampling of pesticides undertaken in Sweden is that of the national agricultural–aquatic environmental pesticide monitoring program of the Swedish University of Agricultural Sciences (SLU), encompassing sixteen shallow wells in four agricultural reference areas. The deficiency of comprehensive monitoring potentially hinders not just sound quality and status assessment, but also the construction, implementation, and evaluation of remedial efforts where they are needed.

The aim of this study is to explore the character and causal conditions of pesticide pollution of Scanian groundwater resources, and from that address, related management and policy issues applying to the wider, national, and general scale. We conduct an in-depth analysis of 23 municipal PSWs, studying their pesticide content as far back in time as possible, and in relation to land use and hydrogeological contexts. Specifically, the following questions are addressed:

- What is the extent and character of pesticide pollution of the investigated groundwater systems as inferred via the PSWs?
- Are there indications of differences in the degree and character of pollution between different types of aquifers and if so, what is generating these differences?

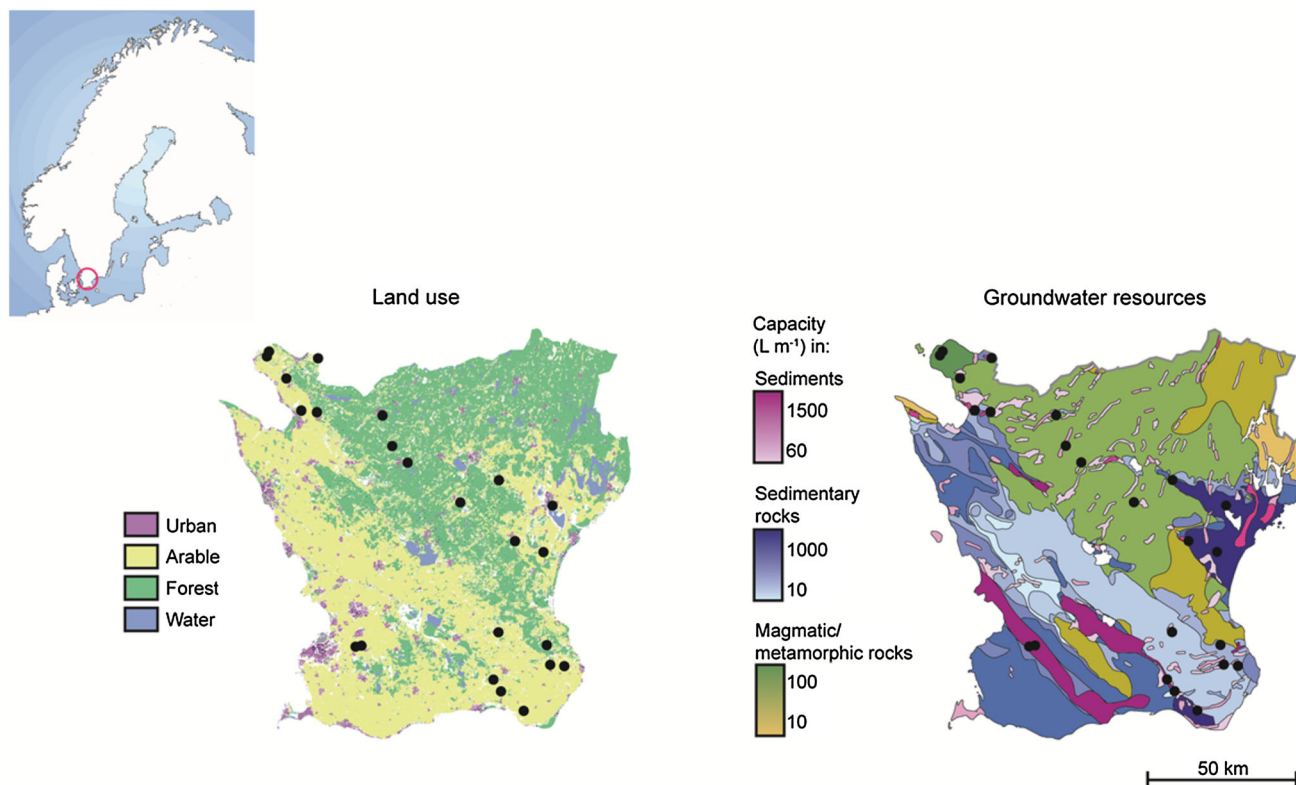


Fig. 1 Location of study area (encircled in the top left-hand corner), associated land use (©Lantmäteriet [I2012/00927]), and overview of regional groundwater resources (Wastenson et al. 1999). Black dots mark the locations of the studied PSWs, for which exact locations cannot be revealed due to non-disclosure agreements with the owners

- Are existing monitoring practices sufficient to provide a sound knowledge basis for status and trend analysis? If not, how can they be improved?

It may be argued that PSWs can be unrepresentative of regional pollution status, as groundwater resources associated with such wells tend to be subject to safeguarding via local regulations on use of potentially hazardous substances and so reflect comparatively less polluted settings. Further, PSWs are generally not ideal for deciphering trends as they generally extract groundwater through long filter screens and hence mix and integrate water of varying origin and quality. Nevertheless, most existing analytical data on pesticide occurrence in groundwater in Sweden stem from such wells, and hence it is both inevitable and relevant to use and study this data for the specified purpose.

MATERIALS AND METHODS

Study area

Scania encompasses roughly 11000 km² at the tip of the Scandinavian Peninsula (55°48'N, 13°37'E; Fig. 1), and experiences Sweden's mildest climate with an average

annual temperature of about 8°C. Precipitation varies between 500 and 1000 mm year⁻¹, depending on coastal proximity and topography (SMHI 2014). Effective precipitation varies between 150 and 500 mm year⁻¹ (Gustafsson 2005).

Compared to other parts of Sweden, Scania comprises relatively little forested land, and is rather characterized by extensive and highly fertile lands, which cover almost half of the region's total area, and maintains much of Sweden's agricultural and horticultural production (SCB 2010). Main crops include cereals, oilseeds, sugar beet, potatoes, and fodder. Pesticide use is common practice throughout the agricultural sector, with a current annual active pesticide ingredient application intensity of about 1.5 kg ha⁻¹ dominated by herbicides, fungicides, and insecticides (in that order, Sandberg 2011). Commonly applied active ingredients throughout past decades include bentazone, isoproturon, metazachlor, simazine, and phenoxy acids such as 2,4-dichlorophenoxyacetic acid (2,4-D), dichlorprop, mecoprop, and MCPA. Urban developments cover about 10% of the total area.

Scania's geological record spans thousands of millions of years and comprises Precambrian crystalline bedrock, various younger sedimentary rocks, and sediment deposits

Table 1 Characteristics of studied wells: *filter depth* (in meters below ground surface; m b.g.s.), *water age group*, *redox state*, *aquifer confinement* and *type*, *land use*, and whether (Y) or not (N) the specific aquifer is associated with a *water protection area*

Well	Filter depth	Water age group	Redox state ^a	Aquifer confinement	Aquifer type	Land use	Water protection area (since)
1	2–54	Modern	Oxic	Unconfined	Fracture	Arable, urban	Y (1974)
2	15–114	Modern	Suboxic	Unconfined	Fracture	Arable, urban	Y (1974)
3	18–22	Modern	Oxic	Unconfined	Pore	Arable, urban	Y (1974)
4	28–40	Modern	Oxic	Leaky	Pore	Arable, urban	Y (1978)
5	6–11	Modern	Oxic	Unconfined	Pore	–	Y (1976)
6	11–23	Modern	Oxic	Leaky	Pore	Arable, urban	Y (2000)
7	74–75	Old	Anoxic	Confined	Pore	Arable, urban	Y (1977)
8	71–72	Old	Anoxic	Confined	Pore	Arable, urban	Y (1977)
9	13–17	Modern	Anoxic	Unconfined	Pore	Arable	N
10	9–12	Modern	Suboxic	Unconfined	Pore	Arable, urban	Y (1977)
11	9–12	Modern	Suboxic	Unconfined	Pore	Arable, urban	Y (2008)
12	5–90	Modern	Suboxic	Unconfined	Fracture	Arable, urban	Y (1980)
13	6–10	Modern	Oxic	Unconfined	Pore	Arable, urban	Y (1973)
14	29–43	Modern	Anoxic	Leaky	Pore	Arable	Y (1987)
15	29–39	Modern	Anoxic	Leaky	Pore	Arable	Y (1990)
16	20–27	Modern	Oxic	Unconfined	Dual porosity	Arable	Y (2008)
17	14–20	Modern	Oxic	Leaky	Pore	Arable, urban	Y (1971)
18	49–69	Old	Oxic	Leaky	Dual porosity	Arable, urban	Y (1980)
19	47–96	Old	Oxic	Leaky	Dual porosity	Arable, urban	Y (1973)
20	14–19	Modern	Oxic	Unconfined	Pore	Urban	Y (1970)
21	9–12	Modern	Suboxic	Unconfined	Pore	Arable	Y (1977)
22	?–57	Modern	Anoxic	Unconfined	Fracture	Arable, urban	Y (1991)
23	7–10	Modern	Oxic	Unconfined	Pore	Arable, urban	Y (1989)

^a Classification according to SGU (2013b). Oxic = class 1, suboxic = class 2, anoxic = class 3

associated with previously glaciated settings. Accordingly, groundwater reservoirs are manifold, spanning fractured gneisses and sandstones, dual-porosity lime and sandstones, and a wealth of sorted, porous, unconsolidated sediment deposits of, primarily, glaciofluvial provenance (Fig. 1; Gustafsson 2005).

Basically, Scania comprises most types of land uses and hydrogeological settings comprised within Sweden as a whole, and accordingly is an interesting area for assessments of both regional and potential national pollution patterns and concerns.

Data collection

The studied PSWs (Fig. 1; Table 1) were chosen after consultation with all Scanian municipalities, the Regional Pesticide Database (RPD), and the Groundwater Bodies, and Water Sources Database (DGV). Selection criteria included overall spatial and hydrogeological distribution, availability of past pesticide analysis records, well meta-data availability, and production importance and access. Approval from well owners was a further requirement.

Results of past pesticide analyses were compiled from drinking water producers, municipalities, the County Administrative Board, relevant commercial laboratories, RPD and DGV, and digitized together with information on analyzed substances and corresponding reporting limits per analysis. The hydrogeological context of each well was determined through studies of geological maps and hydrogeological reports provided by the SGU and the respective local authorities. All wells were classified in terms of aquifer confinement (unconfined, leaky, or confined) and type (pore, fracture, or dual porosity). The presence or absence of arable land and urban areas within the expected recharge area was determined through the Swedish mapping, cadastral, and land registration authority’s terrain map (1:50 000). Whether or not the studied aquifers were associated with water protection areas, i.e., judicial protection areas for which environmentally hazardous activities (generally including pesticide use) are controlled to safeguard potable water supply, was determined via the Swedish Environmental Protection Agency’s VicNatur-database.

In January 2012, all wells were sampled for metals, nutrients, pesticides, and tritium–helium-3 (³H–³He) for

groundwater age assessment. Permanently installed pumping-equipment was used throughout. As active production wells, all had been thoroughly flushed upon sampling. Nevertheless, samples were first retrieved upon field parameter (pH, conductivity, DO) stabilization, as measured in a flow-through cell connected to an Aquameter AP-800.

Metal and nutrient samples were collected from the tap in plastic bottles provided by the analytical laboratory, ALS Scandinavia. Samples were chilled upon retrieval. Fe, Mn, and SO₄ concentrations were used for redox classification, according to SGU (2013b).

Pesticide samples were collected from the tap in glass bottles provided by the Organic Risk Pollutants Laboratory, SLU. Samples were chilled upon retrieval, immediately forwarded to the laboratory and screened for 101 types of pesticide residues (including metabolites) using LC–MS/MS according to Jansson and Kreuger (2010). The median limit of detection (LOD) was 0.003 µg L⁻¹, and the median limit of quantification (LOQ) was 0.01 µg L⁻¹.

Samples for ³H analysis were collected in duplicate 1 L glass bottles, and samples for He isotope analysis in duplicate clamped-off copper tubes connected to the well tap through a closed hose system. Subsequent analyses were carried out at the Bremen Mass Spectrometric Facility according to Sültenfuss et al. (2009). Sampled waters were classified as either (predominantly) modern or old according to ³H concentrations, and after testing for large-scale mixing via comparing initial-³H to historical records of ³H local concentrations in precipitation during year-of-recharge as suggested by the radiometric ³H–³He age (Aeschbach-Hertig et al. 1998). ³H–³He analysis results are specified in Electronic Supplementary Material, Table S1.

Self-organizing maps

Data patterns and variable relations were assessed using Kohonen's self-organizing maps (SOM) approach (Kohonen 2001). The SOM is an unsupervised, exploratory, neural network analysis approach that finds natural populations and patterns within multi-dimensional data sets and allows relationships to be displayed visually. Input samples are compared based on variable properties, then sorted, and mapped onto a set of color-coded 2D graphs (maps) so that similar samples are mapped onto "nodes" with similar characteristics, and nodes with similar characteristics are positioned close to each other on the "map". Both sample and variable similarities and patterns can be assessed through these maps, by the naked eye, and by traditional statistical processing procedures, such as correlation, principal component, and clustering analysis. The SOM handles categorical, continuous, and missing data, and non-linear relationships between variables. The SOM approach is, therefore, suited for the analysis of often complex and

Table 2 Variables included in the SOM analysis. DF 0.01 = detection frequency for the 0.01 µg L⁻¹-threshold data set. DF 0.1 = detection frequency for the 0.1 µg L⁻¹-threshold data set (see "Materials and methods" section)

Category	Variable	Type	
Pesticide pollution degree	DF 0.01	Continuous	(%)
	DF 0.1	Continuous	(%)
Well filter	Top	Continuous	(m b.g.s.)
	Bottom	Continuous	(m b.g.s.)
Water age group	Modern	Categorical	(1/0)
Redox state	Oxic	Categorical	(1/0)
	Suboxic	Categorical	(1/0)
	Anoxic	Categorical	(1/0)
Aquifer confinement	Unconfined	Categorical	(1/0)
	Leaky	Categorical	(1/0)
	Confined	Categorical	(1/0)
Aquifer type	Pore	Categorical	(1/0)
	Fracture	Categorical	(1/0)
	Dual porosity	Categorical	(1/0)
Land use	NO ₃ ^a	Continuous	(mg L ⁻¹)
	Urban	Categorical	(1/0)

^a Arable land is present in all but two of the expected recharge areas (Table 1) inferring a limited basis for statistical analysis of influence based on presence/absence data. Instead, NO₃ concentrations were used as a proxy for agricultural land-use influence

disparate geoscientific data (Fraser and Dickson 2007). The potential of the technique for hydrologic system analysis has been reviewed and concluded upon by Kalteh et al. (2008), with recent examples including, e.g., investigations of heavy rainfall patterns (Nishiyama et al. 2007) and groundwater exploration (Friedel et al. 2012).

The SiroSOM software developed by the Australian Commonwealth Scientific and Industrial Research Organisation (CSIRO) was used for the analysis. Included variables are listed in Table 2. After various trials, we found that 1365 iterations (65 for rough training, 1300 for fine training) onto a 9 × 6 sized (54 nodes) toroidal map, with a hexagonal lattice, was optimum for displaying the variability within the data set. Interpretation was augmented by the use of correlation and principal component analyses of the SOM output.

Due to inconsistencies between compiled pesticide analyses, *for the SOM only*, collected pesticide data were adjusted with respect to analytical boundary conditions to enable comparative studies between wells. The adjustment was conducted with regard to *analyzed substances* so that the data set would only include analyses covering the same residues, and *reporting limits* so that the meaning of detection would be equal for all included analyses. Adjustments were implemented to preserve as much of the data as possible, resulting in an adjusted data set restricted

Table 3 Detected substances with information on *type* (H = herbicide, F = fungicide, I = insecticide), *detection frequency* in %, *no. of wells affected*, *median concentration* (c) and range of documented detections in $\mu\text{g L}^{-1}$, *median LOQ* (limit of quantification), and range of documented analyses in $\mu\text{g L}^{-1}$, *main use* (A = agricultural, Non-A = non-agricultural), and *year of prohibition* (if applicable)

Substance	Type	Detection frequency (incl. trace detections)	No. of wells affected	Median c. (range)	Median LOQ (range)	Main use	Prohibited since
Atrazine	H	6.4 (9.2)	12	0.008 (0.002–0.06)	0.01 (0.001–0.1)	Non-A	1989
Atrazine-desethyl	“	6.3 (10)	10	0.03 (0.01–0.2)	0.01 (0.002–0.2)	“	“
Atrazine-desethyl-desisopropyl	“	4.8 (4.8)	2	0.02 (0.01–0.02)	0.01	“	“
BAM	H	24 (29)	12	0.05 (0.01–0.6)	0.01 (0.003–0.1)	Non-A	1990
Bentazone	H	14 (15)	8	0.09 (0.01–0.4)	0.01 (0.003–0.1)	A	–
Carbendazim	F	2.4 (2.4)	1	0.01	0.002 (0.002–0.01)	A	1998
Clopyralid	H	0.61 (0.61)	1	0.1	0.1 (0.01–0.3)	A	–
Imazapyr	H	1.7 (1.7)	1	0.01	0.01 (0.01–0.1)	Non-A	2002
Imidacloprid	I	4.3 (4.3)	1	0.07	0.002	Both	–
Isoproturon	H	0.84 (0.84)	1	0.2 (0.1–0.2)	0.01 (0.001–0.2)	A	2012
Mecoprop	H	2.4 (2.8)	1	0.01	0.01 (0.003–0.1)	Both ^a	–
Metalaxyl	F	0 (5.3)	2		0.001 (0.001–0.3)	A	–
Quinmerac	H	0.43 (0.87)	2	0.1	0.01 (0.001–0.1)	A	–
Simazine	H	0 (0.41)	1		0.01 (0.001–0.2)	Non-A	1994
Terbuthylazine	H	1.6 (1.6)	2	0.02 (0.002–0.03)	0.01 (0.001–0.1)	Both	2003
Terbuthylazine-desethyl	“	6.4 (11)	5	0.005 (0.003–0.005)	0.01 (0.001–0.01)	“	“
Terbuthylazine-hydroxy	“	17 (17)	1	0.04	0.01	“	“

^a Agricultural use prohibited since 2011

to analyses covering the twenty most commonly analyzed substances (Electronic Supplementary Material, Table S2). A common detection threshold of $0.01 \mu\text{g L}^{-1}$ was applied so that for substances with a lower specified analytical detection limit, concentrations less than $0.01 \mu\text{g L}^{-1}$ were set to 0 (see e.g., Kolpin et al. 1995). A 2nd adjusted pesticide data set was created with a common detection threshold set to the environmental threshold criteria of the EC; $0.1 \mu\text{g L}^{-1}$ (European Union 2006). Subsequently, DFs were calculated per well for both data sets.

RESULTS

Pesticide occurrence and analysis records

Altogether, 255 pesticide analyses were collated from the various sources noted in the methodology section, including our own analyses. Considerable discrepancies between the different data sources were noted both in terms of data quantity and type of data stored. No single source provided a complete record. The frequency and analytical scope of conducted analyses varied between both wells and for single wells over time. The overall sampling frequency per well averaged just below 1 year^{-1} , but varied between 0.4 and 2.4. The number of analyzed substances ranged between 1 and 122 per analysis. LOQ-variation spanned

between 0.001 and $5 \mu\text{g L}^{-1}$. Some wells demonstrated analysis records covering just over two decades, others only covered the past few years.

In total and when including trace amounts, pesticides can be concluded to have occurred in 18 of the 23 wells, i.e., approx. 80 % of the study sites. If considering above-LOQ detections only, the figure is reduced to about 60 % (14 wells). Detected substances are listed in Table 3. Atrazine (and metabolites), BAM, bentazone, and terbuthylazine (and metabolites) represent the most common substances both with regard to overall DF and in terms of number of wells affected (Table 3). The remaining substances listed have been detected once or more in one or two wells only. Over half of the detected substances stem from (i) prohibited compounds, and (ii) compounds mainly or partly used outside of the agricultural sector. Of the four most common substances, only bentazone is (i) currently granted usage and (ii) used solely for agricultural purposes. Metabolites account for more than half of the total number of detections.

DFs for polluted wells range between 13 and 100 %, with an average of about 60 %. Single, isolated detections are rare, suggesting that if a pesticide is detected in a well at a given point in time, repeated pesticide detections in the same well can be expected. More than one type of substance further tends to be detected in a given polluted well throughout its analysis record, suggesting that a well in

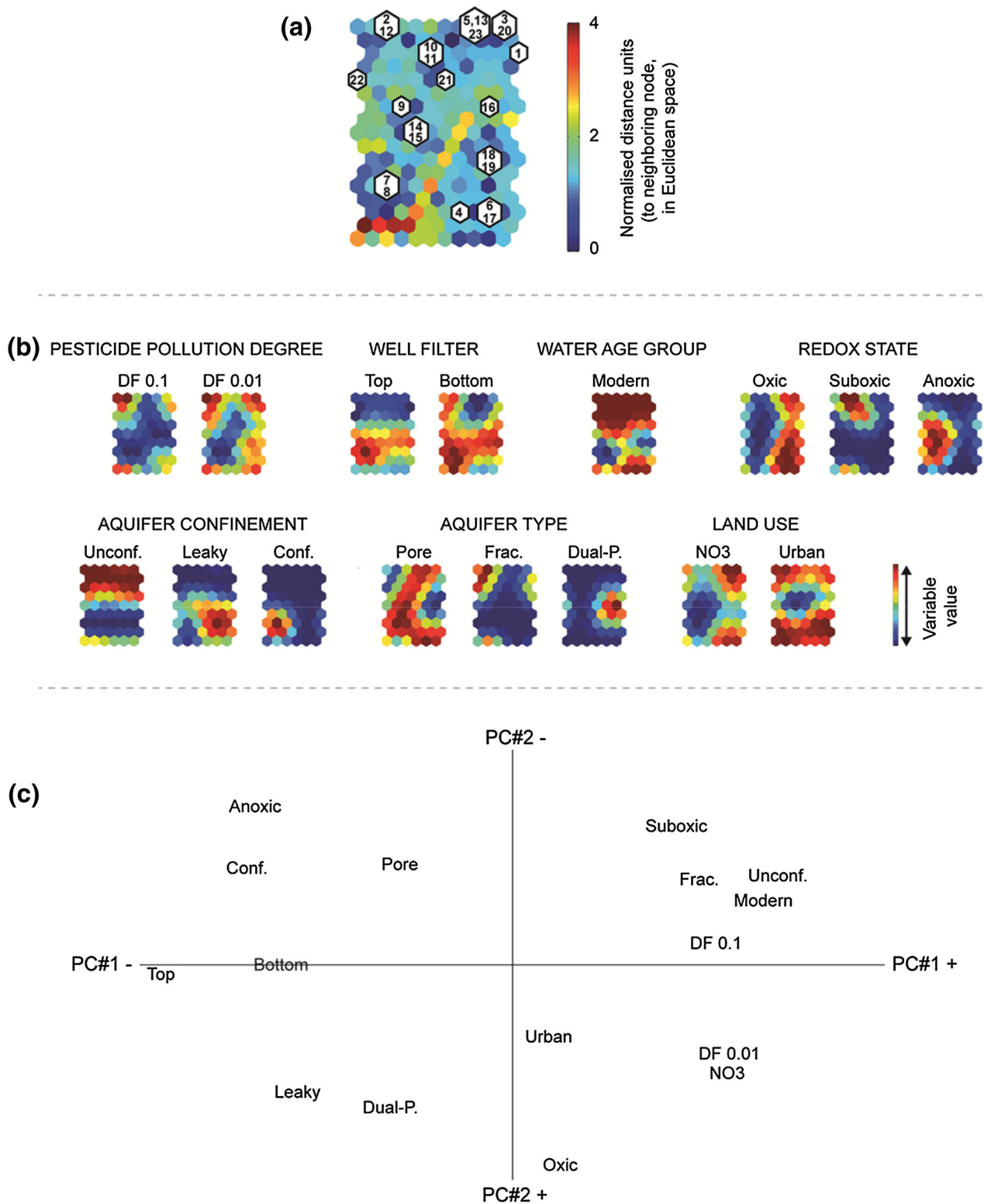


Fig. 2 SOM outputs. Maps are toroidal and so are wrapped top-to-bottom and side-to-side. Note the different color schemes. **a** The U-matrix. Input sample locations in the map are represented by the (well no.) annotated *white nodes* with *black encircling* (the bigger the node, the more input samples represented by it). *Colors* represent degree of dissimilarity/similarity: *warm colors* equal larger distance and dissimilarity, *cooler colors* equal smaller distance and similarity. **b** Component plots, sorted according to variable category, demonstrating the contribution of each variable to the composite U-matrix and hence variable variation over the data space. *Warm colors* correspond to high values (for binary variables; 1), *cool colors* correspond to low values (for binary variables; 0). Similar patterns between variables indicate positive correlation, inverse patterns indicate inverse correlation. **c** A “similarity plot” based on a principal component analysis of SOM component plots (b). Proximity indicates similarity

Table 4 SOM component plot (Spearman rho-) correlation analysis results (hit nodes only). Bolded *r*-values are significant at >90 %. Bolded and underlined *r*-values are significant at >95 %

Category	Variable	DF 0.01 (<i>r</i>)	DF 0.1 (<i>r</i>)
Well filter	Top	<u>−0.604</u>	<u>−0.609</u>
	Bottom	−0.009	0.125
Water age group	Modern	0.351	0.504
Redox state	Oxic	<u>0.551</u>	0.149
	Suboxic	−0.079	0.193
	Anoxic	<u>−0.559</u>	−0.325
Aquifer confinement	Unconfined	0.384	0.371
	Leaky	−0.109	−0.194
	Confined	<u>−0.540</u>	−0.375
Aquifer type	Pore	−0.478	−0.317
	Fracture	<u>0.566</u>	<u>0.741</u>
	Dual porosity	0.015	−0.379
Land use	NO ₃	<u>0.656</u>	0.412
	Urban	<u>0.545</u>	0.469

which one type of pesticide is detected at a given point in time, is likely at risk of being polluted by additional types of substances. However, there are wells in which only single substances are detected repeatedly. Recurrent substances in such wells include BAM, bentazone, and mecoprop.

Documented concentrations of individual substances range between estimated trace amounts of 0.001 µg L^{−1}, to measured concentrations of up to 0.64 µg L^{−1}. Concentrations above the EU-threshold of 0.1 µg L^{−1} for a single substance have been measured one or more times in nine wells. Bentazone is the substance most commonly detected in above-threshold concentrations. Atrazine-desethyl, BAM, clopyralid, isoproturon, and quinmerac, which are all relatively mobile herbicides, have also been detected in concentrations ≥0.1 µg L^{−1}.

The results of the comprehensive low-LOQ-analyses conducted as part of this study are presented in Electronic Supplementary Material, Table S3. These revealed (i) pesticides in three wells wherein pesticides had not been detected previously, (ii) for the specific well previously undetected pesticides in fifteen wells, and (iii) three pesticides which had never been detected before in any of the wells. The latter three included imidacloprid (1 well), simazine (1 well), and metalaxyl (2 wells). Simazine, a herbicide, has been prohibited for use in Sweden since 1994, while imidacloprid (insecticide), and metalaxyl (fungicide) are currently allowed. Only metalaxyl is exclusively used within the agricultural sector.

The variability in analytical scope between samples and wells, combined with the uncertainty relating to (i) potential offsets between groundwater and pesticide

travel times, and (ii) representativeness of derived groundwater ages in view of expected groundwater mixing, currently precludes trend analyses both regionally (between wells) and locally (for an individual well).

SOM-results: data patterns and variable relationships

SOM-outputs are given in Fig. 2a–c and Table 4.

Figure 2a is the “Unified distance matrix” (U-matrix—Ultsch and Siemon 1990; Ultsch 2004) representation of the self-organized map. It is an overall and composite visualization of the SOM that displays nodes grouped or distributed across the map according to overall similarity. The white nodes are so-called “hit nodes” representative of the location of actual input samples within the data space. The size of a hit node is proportional to the number of input samples represented by it. Fourteen hit nodes were sufficient to represent the variability of the 23 input samples within the given data space. Dissimilarity/similarity between neighboring nodes is indicated via color-coding; the cooler the color of a node, the greater the similarity to surrounding nodes (and vice versa).

Figure 2b shows the component (plane) plots derived from the SOM analysis. These plots are basically slices of the self-organized map, showing each variable’s contribution across it. The location of input samples is the same as for the U-matrix as both are derived from the same SOM analysis. On the component plots, cool colors (blue) represent low variable values, and warm colors (red) represent high variable values. Coinciding patterns between two or more variables indicate positive correlation (such as between DF 0.1 and DF 0.01), and inverse patterns indicate inverse correlation (such as between filter top depth and modern water). Results of image processing procedures applied to the component plots are given in Fig. 2c (principal component analysis; PCA conducted on all nodes) and Table 4 (correlation analysis conducted on hit nodes only).

The patterns of the component plots (Fig. 2b) indicate that severe, high concentration pesticide pollution (the elevated yellow–red regions of the DF 0.1 plot) is related to modern, oxic–suboxic waters with high NO₃ concentrations from wells with relatively shallow filter tops in unconfined fracture or pore-type aquifers exhibiting urban land-use influence. The PCA (Fig. 2c) and the correlation analysis (Table 4) expectedly corroborate these indications, and further suggest particular strong associations between DF 0.1, modern water, fracture-type aquifers, urban land use and filter top depth (such that the shallower the filter top, the more polluted the water).

Similarly, the least polluted waters (represented by the depressed blue region on the DF 0.01 plot) appear to be

primarily associated with deeper and more confined groundwater environments, suboxic–anoxic redox states, pore-type aquifers, and low NO_3 concentrations (Fig. 2b, c; Table 4). Because of correlation between anoxic conditions and low NO_3 concentrations, it is difficult to interpret the latter as an actual sign of low agricultural influence, as it is rather likely to primarily relate to nitrate reduction. Interestingly, there is no convincing correlation between overall pesticide presence/absence and predominantly modern/old groundwater. Arguably, some of the so classified old waters still contain a large enough proportion of modern water to allow for pesticide detection, albeit not for high concentration pesticide presence.

DISCUSSION

Pollution extent, character and vulnerability

Repeated pesticide occurrence applies to a majority of the studied PSWs. Because PSWs generally integrate water quality over large recharge areas, pesticide occurrence in these types of wells is likely to reflect either large-scale pollution in terms of volume, or in terms of source-concentrations (Brüsch 2007). All but one of the studied aquifers are further subjected to regulated safeguarding measures (Table 1), whereby these may effectively be non- (i.e., under-) representative of general conditions.

On the other hand, as the PSWs were chosen partly based on pesticide analysis record length, there is potential for bias toward the relatively more polluted examples of regional groundwater reservoirs used for drinking water production, as these in the absence of a general sampling prerequisite, may have demanded relatively more attention than others. This could be a valid reasoning as previous studies have indicated lower pesticide DFs, in the range of 30–50 %, for Scanian groundwater reservoirs (Maxe et al. 2003; Virgin 2012), as compared to the derived 80 % of this study. However, it is difficult to compare these numbers directly in view of varying investigative scope and approach, and this study arguably allows for a relatively more thorough assessment in view of the detailed analysis records compiled (including conducted low-LOQ analyses). Either way, pesticide pollution of Scanian groundwater seems a genuine concern on par with established situations in many other industrial countries, in terms of overall DF, concentration levels, and frequently occurring substances (Kolpin et al. 1998; Haarstad and Ludvigsen 2007; Schipper et al. 2008; Close and Skinner 2012; Thorling et al. 2012), and questions the previous reports of no danger of pesticide pollution of groundwater for Sweden as a whole (European Environment Agency 2004).

While agriculture is undoubtedly an important source of pesticide pollution of the sampled waters, the collated data clearly indicate that non-agricultural pesticide use also exerts a major influence. As noted earlier, similar indications have been demonstrated also in other regions including e.g., the US (Kolpin et al. 1998) and Denmark (Thorling et al. 2012). One could speculate whether the major urban influence might result from a relative lack of restrictions in terms of non-agricultural compared to agricultural pesticide use. In Sweden, however, large efforts have been put into the restriction of non-agricultural pesticide use since the end of the 1980s, when studies started to indicate significant environmental impact thereof. Potent weed killers such as atrazine and dichlobenil (the parent compound of BAM) were effectively prohibited for use by 1990, and thus many of the observations made likely stem from a time during which overall restrictions were absent or less strongly enforced. One part of the explanation for the extensive occurrence of these types of substances in the studied aquifers is certainly inherent and generally valid hydrogeological system time lags (e.g., Destouni et al. 2010). Gradual leach-out of built-up pools of recalcitrant substances within the soil could be yet another concern. In comparable hydrogeological settings in Denmark, BAM is recognized as a potential long-term groundwater pollution threat in view of documented soil loading (Clausen et al. 2007). Similar indications have been made in relation to atrazine and associated metabolites in e.g., Germany (Tappe et al. 2002) and France (Morvan et al. 2006). No comparable studies are known from Sweden.

In terms of pollution vulnerability, the results confirm the expected importance of connectivity between the ground surface and the point of observation within the groundwater system. According to the SOM output, vulnerability and thus pollution risk generally increases with modern and oxic waters, with decreasing minimum extraction depth, and with decreasing degree of aquifer confinement. These findings are typical (e.g., Close and Skinner 2012; Malaguerra et al. 2012), expected, and in agreement with the long-standing general consensus (Barbash and Resek 1996). Indications are further that groundwater systems dominated by fracture flow are particularly vulnerable to pesticide pollution. Likely, fracture flow increases pollution vulnerability via an overall reduction in both travel time and natural remediation-potential relative to matrix flow, thereby allowing for comparatively rapid transfer of infiltrating water and pesticides. This proposal is theoretically valid (Freeze and Cherry 1979) and well-established regarding fracture and macropore flow, and associated pollutant transport in e.g., clay tills (Jorgensen et al. 1998; Jarvis 2007). However, few studies comparing pesticide pollution in aquifers

dominated by matrix flow to those dominated by fracture flow have been able to support such a hypothesis (Neil et al. 1989; Barbash and Resek 1996). Crystalline fracture-type aquifers are present throughout much of Sweden (and much of Scandinavia), and hence, this pollution concern is in need of further investigation.

Finally, it is interesting to note that a majority of the studied aquifers are polluted even though all but one have water protection areas. Many of these areas were installed in the mid-1970s and so should have served to protect the associated groundwater quality since then. Thus, there appears to be a need for re-evaluation of (i) the location of the areas in relation to actual recharge areas, and (ii) the scope of associated regulations.

Data and management limitations and requirements

While this study fills an important role in indicating regional concerns of pesticide pollution of groundwater and the causes of those, the underlying data set is limited which constrains possibilities for confident, general conclusions. Whether or not the above inferences are valid also in a wider context requires extended studies encompassing more wells and desirably also more comprehensive and detailed metadata concerning e.g., land use patterns over time and space within well-defined recharge areas, and groundwater age distributions of extracted waters. The lack of coordinated pesticide-groundwater monitoring, as well as the lack of a comprehensive database of conducted analyses, is a palpable concern in this regard, as it constrains possibilities for rigorous status assessments over both space and time. Regional temporal and spatial comparisons currently require data collation from a wide range of separate authorities and databases followed by data quality assessment and leveling. This approach is neither efficient nor ideal as it is excessively time-consuming and jeopardizes objectivity. The limited availability of well and sample metadata including information on e.g., filter depths, recharge areas, and stratigraphy is a further concern.

For now, an effort to collate and structure all documented pesticide analyses of groundwater in Scania and overall Sweden from the variety of information sources to a single master database should be made, to enable a wider test of the hypotheses put forward here, and to further assess the current pollution situation. Such a database needs to contain fundamental well and sample metadata, and should be continuously maintained and updated. Requirements for automated submission of conducted analyses of pesticides in groundwater, inclusive of analytical boundary conditions and specified metadata, to this database should be implemented. Further, multiple catchment-scale field studies should be undertaken in various

type settings to study governing transport processes in detail (e.g., Åkesson et al. 2013), to further explore the extent and effects of system time lags on pesticide fate in the groundwater system (e.g., Tesoriero et al. 2007), and to investigate the potential presence, extent, and effects of pesticide residues within soils. Such studies are fundamental both in terms of enhancing pollution prediction skill, and for the development of sound management policies including remedial efforts where needed.

In order to secure WFD compliance and thereby also future groundwater quality, Sweden further needs to develop and implement statutory regulations enforcing adequate monitoring and assessment of the occurrence of not only pesticides but, as previously noted, also other anthropogenic pollutants (Destouni et al. 2008; Baresel and Destouni 2009; European Commission 2012; SEPA 2013) in groundwater nationwide. As for general groundwater quality monitoring and assessment, precepts effectively already exist (e.g. SGU 2013:1). However, these lack distinct pesticide monitoring and assessment specifications, and appear poorly followed overall. Based on experiences from neighboring Denmark, a country well ahead of Sweden as to this matter (Jorgensen and Stockmarr 2009), adequate monitoring, and assessment should span both untreated groundwater from PSWs, and groundwater from an independent and comprehensive purpose-made monitoring network, in order to secure a sustained, geographically, and hydrogeologically well-distributed, controlled source of data.

CONCLUSIONS

- Analysis records from 23 public supply wells reveal pesticide occurrence in all but five, which suggests that pesticides are substantially present in groundwater throughout much of Scania, southern Sweden. Collected data indicate considerable influence of both agricultural and non-agricultural (urban) pesticide use, and lagged effects of past, less restricted pesticide use. Modern (post-1950), oxic waters from shallow, unconfined, unconsolidated or fracture-type bedrock aquifers appear particularly affected and vulnerable. Least affected waters appear primarily associated with relatively deeper extraction depths, anoxic conditions, and more confined sediment aquifers largely devoid of urban land-use influence. Predominantly old (pre-1950) water does not appear to exclude pollution risk, which could be due to earlier pesticide use, and/or relatively minor proportions of modern, severely polluted water.
- Pesticide residues that had not previously been observed via more standard analyses were detected throughout many of the studied aquifers when applying

a comparatively generous screening analysis package covering an ample range of residues detectable at relatively low concentrations. This implies that the standard analysis packages are not sufficient for thorough pesticide–groundwater pollution status assessment.

- As a majority of the studied aquifers are affected by pesticide pollution even though all but one are associated with water protection areas, the positioning of and pesticide use regulations associated with these areas should be reassessed.
- Due to the relatively limited data set explored, further studies are required to test the above inferences of primary pollution concerns in more detail and in a wider context. Such studies are currently hampered by a lack of comprehensive and consistent data on pesticide occurrence in Swedish groundwater, resulting from nationally deficient statutory monitoring of anthropogenic pollutants in groundwater (Destouni et al. 2008; Baresel and Destouni 2009).
- For Sweden, comprehensive monitoring of the occurrence of pesticides (and other anthropogenic pollutants) in groundwater needs to be realized, and available data need to be collated and structured to allow for comprehensive nationwide status assessment, identify problem areas, direct research efforts, and develop and implement sound management plans in accordance with the WFD.

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