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Greenhouse production contributes to pesticide occurrences in Swedish streams

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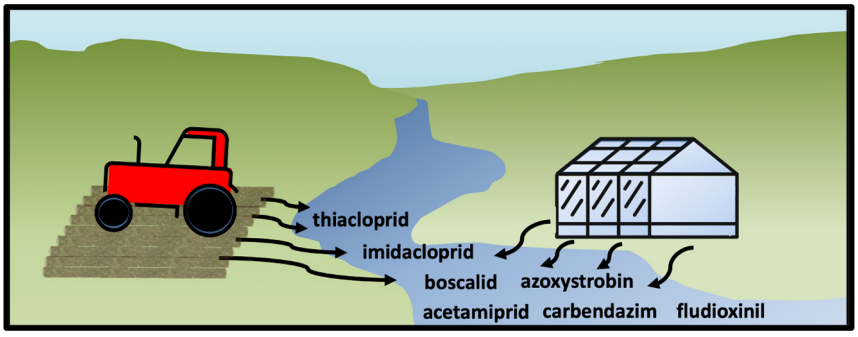
HIGHLIGHTS

- All measured pesticides used in greenhouses were detected in recipient streams.
- Frequently used substances most prone to elevated downstream concentrations.
- Imidacloprid most frequently detected at potentially ecotoxicological concentrations.
- Higher concentrations of used substances than in catchments without greenhouses.

GRAPHICAL ABSTRACT



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ABSTRACT

Greenhouse and other covered cultivation systems have increased globally over the past several decades, leading to considerably improved product quality and productivity per land area unit. However, there is a paucity in information regarding the environmental impacts of covered production systems, especially regarding pesticides entering the surrounding environment. Aiming to address this knowledge gap, we collected grab samples downstream of greenhouses from seven Swedish streams every 14 days during a 12 month period. In three of the streams, samples were also taken upstream of the greenhouses and in four of the streams time-integrated samples were collected by TIMFIE samplers in the period between grab sampling occasions. The samples were analyzed for 28 substances (27 that were permitted for use in greenhouse production systems in Sweden and one degradation product to a permitted substance). Pesticide use journals were collected from the greenhouse producers for the 12 month period. The results were examined for indications of greenhouse contributions to detection frequencies, maximum and average concentrations, and potential ecotoxicity in several ways: (1) comparing locations downstream of greenhouses with registered use of a substance with those without registered use, (2) comparing results from this study with those from the Swedish environmental monitoring program of pesticides in surface water from catchments with no greenhouses from the same period and region, (3) comparing concentration trends with registered pesticide application times in the greenhouses, and (4) comparing up- and downstream concentrations. The results strongly suggest that greenhouse applications do contribute to

Abbreviations: AF, Assessment Factor; EFSA, European Food Safety Authority; EM, Environmental Monitoring; GH, Greenhouse; LOD, Limit of Detection; LOQ, Limit of Quantification; O, Ornamentals; OMK, Organic Environmental Chemistry Laboratory; PNEC, Predicted No-Effect Concentration; RAR, Renewal Assessment Report; SLU, Swedish University of Agricultural Sciences; SPE, Solid Phase Extraction; SWEDAC, Swedish Board for Accreditation and Conformity Assessment; TIMFIE, Time-Integrating, Micro-Flow, In-line Extraction; TER, Toxicity Exposure Ratio; V, Vegetables.

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pesticide occurrences, maximum and median concentrations for most of the pesticides included in this study, and to potential toxicity to aquatic organisms for several of them, most notably imidacloprid, acetamiprid, carbendazim, and pirimicarb.

1. Introduction

Greenhouse production has increased steadily since its commercial introduction about half a century ago (Boulard et al., 2011; Nordey et al., 2017). Covered production systems are now estimated to represent about half of the total production of fresh vegetables worldwide (Boulard et al., 2011) and a considerable contribution to national economies (White et al., 2019). Covered crop production, including greenhouses, have significantly increased the productivity - both in terms of yield and crop quality (Nordey et al., 2017) – compared to open-field agriculture in terms of per land area unit (Boulard et al., 2011; Majsztrik et al., 2017) and, in many cases, also per water unit (Colino Sueiras and Martínez Paz, 2002; Majsztrik et al., 2017). However, the high intensity of greenhouse production systems has raised concerns about the environmental impacts; mainly regarding the energy consumption associated with temperature control (heating/cooling) and the more frequent and higher doses (per land area unit) of fertilizer applications than in open-field farming (Boulard et al., 2011; Majsztrik et al., 2017). In life-cycle assessments (LCAs) of greenhouse production systems, heating is normally found to be the main contributor to overall environmental impacts (Antón et al., 2004; Boulard et al., 2011; Canakci and Akinci, 2006; Parrado and Bojacá, 2008; Torrellas et al., 2013), with infrastructure and/or fertilizers being reported as the secondary contributors (Bojacá et al., 2014). In un-heated systems, irrigation, infrastructure, fertilizers, and/or agrochemical use – depending on the specific system - leave the largest ecological footprint (Canaj et al., 2020). However, there are large discrepancies in terms of covered production system efficiencies and their environmental impacts in different climatic, socio-economic, and geographical regions (Canaj et al., 2020; Nordey et al., 2017; van Lenteren, 2000). Thus, it remains unclear which system is the most efficient in terms of minimizing overall environmental impacts per unit of product, given the generally high productivity per unit area in greenhouses, the wide variety of covered crop production systems, geographical/climatic/socio-economic differences, and the lack of empirical data to allow for direct comparisons between open-field and covered production, as well as between different covered production systems on a yield-based level.

With regards to pesticides, there is a paucity in empirical studies of the actual environmental impacts from different types of covered production systems, and how they compare to each other and with open-field agriculture, in particular regarding ecotoxicity. LCAs have indicated the overall environmental impact of pesticide use in covered production systems, and especially in soil-less greenhouses, is small compared to other aspects of those production systems (Boulard et al., 2011). However, it should be noted that only a few aspects of pesticide application are typically considered in LCAs, *e.g.,* the energy cost of production and transport of pesticides (Bojacá et al., 2014; Canaj et al., 2020). In general, pest control is considered to be easier to manage in covered systems, due to the ability to monitor and target specific pests and employ integrated pest management (IPM) systems (van Lenteren, 2000), which (at least theoretically) should result in more efficient pest treatment. However, in terms of economics, the costs for chemical pesticides are minimal compared to other greenhouse production costs (van Lenteren, 2000), so the economic drivers to minimize pesticide use are limited.

To date, most empirical studies of unwanted pesticide application effects in greenhouses have focused on the exposure of greenhouse workers during and after application (Bouvier et al., 2006; Hatzilazarou et al., 2004; Negatu et al., 2017; Nuyttens et al., 2009), as well as to florists (Bouvier et al., 2006; Song et al., 2014; Toumi et al., 2017, 2019) and consumers (Bouvier et al., 2006; Leili et al., 2016; Zhang et al., 2015) from pesticide residues on the harvested products. In terms of environmental impacts of pesticides from greenhouses (and other covered production systems), investigations have primarily focused on assessing potential pollution pathways (González-Pradas et al., 2002; Katsoulas et al., 2012), including leaching into the soil below the greenhouse (Garratt et al., 2007; Haarstad et al., 2012; Hatzilazarou et al., 2004) and, by extension, to groundwater (Haarstad et al., 2012; Hatzilazarou et al., 2004), volatilization and drift leading to pesticides in the air (Doan Ngoc et al., 2015; Hatzilazarou et al., 2004), and residues in the leachate draining from the greenhouses (Haarstad et al., 2012; Hatzilazarou et al., 2004). Only a few studies have empirically examined the actual contribution of greenhouses (and other covered production systems) to surface water concentrations of pesticides (Kreuger et al., 2010; Leistra et al., 1984; Roseth and Haarstad, 2010). This is possibly due to the fact that greenhouses have been assumed to prevent release of pesticides to the environment (EU, 2009). However, it has been noted that this is not true for many systems (Hatzilazarou et al., 2004; Vermeulen et al., 2010; Vermeulen et al., 2015; White et al., 2019). Together with the previously mentioned studies indicating that pesticides from greenhouses do indeed enter surface waters (Kreuger et al., 2010; Leistra et al., 1984; Roseth and Haarstad, 2010), this suggests that more attention should be paid to the contributions from greenhouses (and other covered production systems) to the occurrence and potential toxicity of pesticides in surface waters, as well as other environmental compartments (EFSA, 2014a). It is, however, difficult to separate out the contribution of pesticides to surface waters from greenhouse production compared to other potential sources in areas where open-field agriculture and other pesticide-applying land uses (including parks, gardens, turf grass areas *etc.*) are present within the same catchment – many substances are permitted for multiple uses. Nevertheless, it is important to investigate and attempt to account for the various sources of pesticide occurrences in streams and other surface waters, in order to establish effective mitigation measures and best management practices targeting the key sources.

Here, we present results from grab sampling (every 14 days) and parallel time-integrated sampling (over the same 14 day periods) using TIMFIE (Time-Integrating, Micro-Flow, In-line Extraction (Jonsson et al., 2019)) from late June 2017 until early July 2018 (*i.e.* ~12 months of data) in up to seven small streams in southern Sweden with one or more greenhouse producers within the immediate upstream area. The sampling was complemented by obtaining pesticide application journals (dates and products, not amounts) from the greenhouse producers. The stream sampling data was compared with results from the Swedish national environmental monitoring (EM) program of pesticides in surface waters from small agricultural catchments without greenhouse producers within the same climatic/geographical region (Boye et al., 2019). The purpose of the current study was to examine the specific contribution of greenhouse production to pesticide occurrence and potential ecotoxicity in Swedish streams.

2. Materials & methods

2.1. Study areas and pesticide applications

The sampling locations (Table 1) were situated in southern Sweden within agricultural catchments (1–212 km^2) with intensive greenhouse production 15–1600 m upstream of the sampling location (with ranges >600 m only applying to the most upstream greenhouse(s) within catchments containing more than one greenhouse facility). The greenhouse production systems are representative of Swedish greenhouses in general, although the facilities are larger than the average greenhouse facility. All greenhouse producers in this study have several greenhouses within the same facility, typically having been constructed over several decades and having

Table 1
Information about study areas.

Location	Catchment size (km^2)	Type of GH production	# of GH facilities	# of grab (TIMFIE) samples[a]	# of used substances (of which analyzed)	Total # of applications	Most frequently used of analyzed substances[b] (% of total # of GH applications in study area)
O1	212	Ornamentals	1	27 (25)	9 (4)	150	acetamiprid (22), pymetrozin (7), propiconazole (6), pirimicarb (5)
O2	96	Ornamentals	2	27 (25)	17 (8)	92	pyraclostrobin (8), boscalid (8)
O3	7	Ornamentals	1	27 (0)	22 (15)	254	propiconazole (12), thiophanate methyl (6)
OV4	1	Vegetables	2	27 (6)	21 (13)	133	imazalil (7)
		Ornamentals	3				
V5	18	Vegetables	2	27 (25)	11 (8)	75	propamocarb (24), imazalil (13), imidacloprid (11)
V6	15	Vegetables	1	26 (0)	8 (5)	19	propamocarb (16), pymetrozin (11), azoxystrobin (5)
V7	3	Vegetables	2	27 (0)	7 (4)	13	hexythiazox (23), azoxystrobin (15), propamocarb (15), imazalil (8)

[a] From down-stream locations.
[b] Substance applications constituting more than 5% of the total number of applications in greenhouses in the study area.

different growing and watering systems. In total, seven catchments with greenhouses were sampled: three hosted only ornamental greenhouse systems (O1–3), three hosted only vegetable producing greenhouses (V5–7), and one hosted both ornamental and vegetable producing greenhouse facilities (OV4). The ornamental production within the study areas primarily took place on tables and the majority of the drainage water was collected in one or more reservoirs on the property. The vegetables were grown in hanging pipes or in buckets on the floor and all vegetable production facilities had water re-circulation and collection systems with central reservoirs. All the greenhouse facilities, except one, had concrete walkways with the soil surface in between being covered in plastic or ground fabric; the single exception had full concrete flooring throughout the facility. Most greenhouses were placed on previous open-air agricultural fields, which are typically tile-drained in southern Sweden. For privacy reasons we are not able to provide more precise locations or details about the study areas.

Of the 31 substances that were used within the greenhouses during the study period, according to the journals supplied by the producers, 21 were included in the analyses of surface water samples in this study (Fig. 1, SI-Table S1-S2). The substances not included in our analyses are complicated to analyze and would require separate analytical methods that were not available in our laboratory. For example, the two most frequently used substances, growth regulators daminozide and chlormequat chloride, are highly polar and, hence, cannot be analyzed by the LC-MS/MS multi-methods (based on reversed-phase chromatography) used in this study. Five substances (three analyzed: paclobutrazol, pirimicarb, and pyriproxyfen) of the used substances were exclusively permitted for use in greenhouse systems (*i.e.* they were not permitted for open-field production or any other type of use in Sweden during the study period). Ornamental production was continuous throughout the year and these producers used the largest number of different substances and also applied pesticides more frequently than the vegetable producers (production period January to early November). However, this does not necessarily mean that the amounts of pesticides used were larger; it is important to note that the pesticide application journals only include dates and types of pesticides (products and active ingredients), not the amounts.

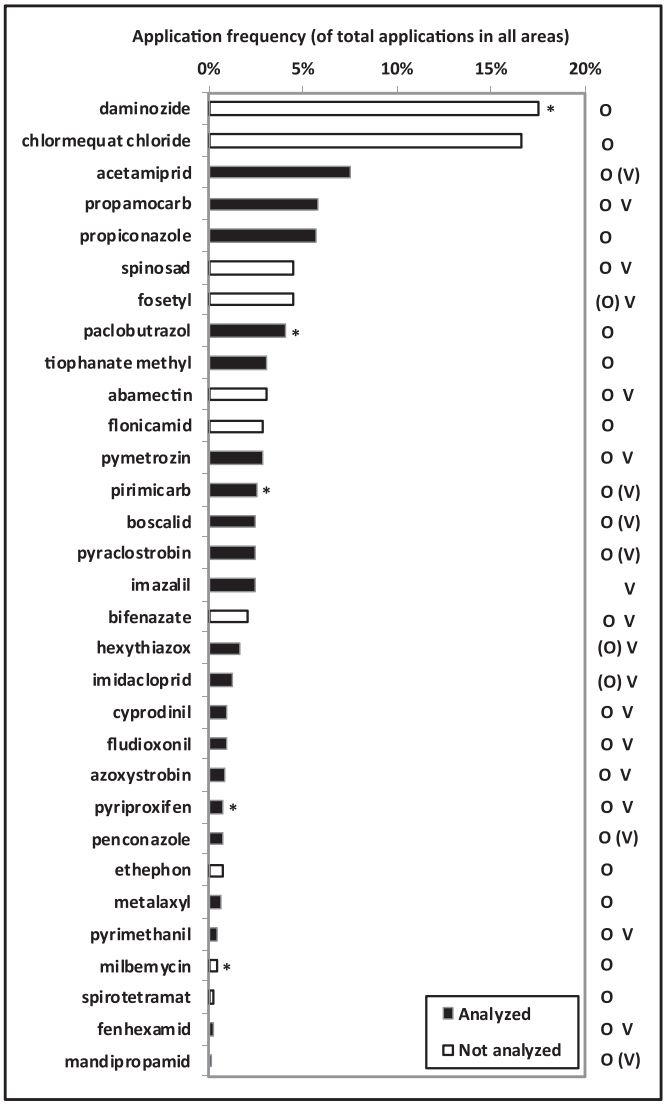

Fig. 1. Application frequency, of the total number of applications in all study areas, for all substances used in one or more of the greenhouses in the study areas during the period sampled. Substances not analyzed within this study are represented by unfilled bars, analyzed substances by filled bars. Substances with an asterisk (*) at the end of the bar were exclusively permitted for greenhouse production during the study period. Letters on the right indicate if the substance was used for O = ornamentals and/or V = vegetables, letters within parantheses indicate the substance was permitted, but not used within the corresponding greenhouse production type in the study areas during the study period.

2.2. Sampling

2.2.1. Grab samples

Grab samples were collected every 14 days, beginning on 26 June, 2017 and finishing on 2 July, 2018, with the exception of a 21-day interval from 12 December 2017 to 2 January 2018. In total 278 grab samples were collected from the seven study areas. In three areas (O1, O2, and V6) grab sampling was conducted both upstream and downstream of the greenhouses. In the other four areas, only downstream (relative to greenhouses) sampling was conducted.

Grab samples downstream of ornamental producers were collected in 1 L glass bottles to allow for analysis of substances specific to ornamental production (method OMK 51, in addition to methods OMK 57 and 58, Table 2). Grab samples collected downstream of vegetable producing

Table 2
Brief description of the analytical methodology used for detection and quantification of total water concentrations of substances in this study.

Method ID	Number of substances[a]	Type of substances	Pretreatment	Extraction/filtration	Detection (ionization)[b]
OMK 51[c]	24 (1)	Non-polar/semi-polar	None	Liquid-liquid extraction dichloromethane	GC–MS (NCI), GC–MS/MS (EI)
OMK 57[d]	109 (27)	Semi-polar/polar	pH adjusted to ~5	Filtration (0.2 μm)[d], online SPE	LC-MS/MS (ESI+)
OMK 58[d]	15 (0)	Acidic semi-polar/polar	Acidification with 1% formic acid to pH ~2.5	Filtration (0.2 μm)[d], online SPE	LC-MS/MS (ESI−)
OMK 60[e]	106 (27)	Semi-polar/polar	None	SPE (during sampling in field), online SPE	LC-MS/MS (ESI+)

[a] Numbers in parantheses indicate numbers of the 27 substances permitted for use in greenhouses (plus thiophanate methyl degradation product, carbendazim) analyzed by each method.
[b] GC–MS = gas chromatography with mass selective detection (mass spectrometry), NCI = negative chemical ionization, GC–MS/MS gas chromatography with tandem mass selective detection, EI = electron ionization, LC-MS/MS = liquid chromatography coupled with tandem mass spectrometry, ESI = electrospray ionization; positive (+) or negative (−).
[c] OMK 51 was only used for O sampling points.
[d] Internal standards are added prior to filtration to account for any losses to the filter and/or particles.
[e] OMK 60 is the method used for TIMFIE samples (detection and ionization methods are identical to OMK 57).

greenhouses were collected in 0.2 L glass bottles, as only two analytical methods (OMK 57 and 58) were employed for those samples. At all locations, the glass bottle was attached to a long rod to enable sampling in the fastest-flowing portion of the stream. After sampling the bottles were shipped (overnight) to the laboratory in insulated boxes on ice.

2.2.2. TIMFIE

Time-integrated sampling using TIMFIE (Jonsson et al., 2019) was conducted downstream of the greenhouses in four of the study areas (O1, O2, OV4, and V5), during the same time period as the grab samples (26 June 2017–2 July 2018), except for in OV4 (April–June 2018). TIMFIE samplers were mounted on telescopic aluminum-rods and positioned at a similar distance away from the shoreline as the grab samples were taken. The TIMFIE samplers are designed to slowly collect water through a narrow PEEK tubing flow restrictor and two serially connected columns: 1. Hydrophobic polymer (Chromafix HR-P, 50 mg, Macherey-Nagel, Düren, Germany) and 2. weak anion exchange (Chromafix XAW, 50 mg, also from Macherey-Nagel) for solid phase extraction (SPE). The water flow is driven by the negative pressure created from a single use plastic syringe with the plunger locked in pulled-out position by a stopper. Thus, no power supply or batteries are needed. Accurate reading of the extracted water volume collected in the syringe barrel enables quantitative determination of target compounds. The maximum sampling time is dependent on the dimensions of the PEEK tubing, which controls the flow rate, and the syringe volume. In this study the samplers were modified to allow for 14-day sampling periods, *i.e.* the samplers were replaced every 14 days and the measured concentrations represent the average for the 14-day sampling period between the grab sampling time points. On average the sampling volume over 14-days was 58 mL (with a relative standard deviation of 32%) for the 90 collected samples. The variability in collection volumes was probably partly dependent on seasonal changes in temperatures, which affects the water viscosity (especially at close-to-freezing temperatures), and hence, flow rate. Another source of variation was rapidly dropping discharge volumes in the streams, which on a few occasions left the samplers above the water surface for parts of the sampling period, in spite of careful positioning of the samplers below the water/ice surface at the time of deployment. The TIMFIE procedure in the analytical laboratory includes adding the internal standard solution to the SPE columns prior to elution of investigated compounds from the sorbent materials and co-extracted suspended particles, thus giving the total water concentration.

2.3. Analyses

2.3.1. Pesticide analyses

All pesticide analyses (methods summarized in Table 2) were carried out at the organic environmental chemistry laboratory (OMK) within the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (SLU). The laboratory regularly participates in international inter-calibration testing and all analytical methods for the grab samples are accredited according to the Swedish Board for Accreditation and Conformity Assessment (SWEDAC). The analytical method for the time integrated, TIMFIE, samples (OMK 60, Table 2), though not yet accredited, has been validated according to similar procedures and principles as the accredited methods. The instrumental part of OMK 60, *i.e.* the liquid chromatography tandem mass spectrometry (LC-MS/MS) with positive electron spray ionization (ESI+) method is the same as the accredited OMK 57 method (Jansson and Kreuger, 2010), which was used for the grab samples. All methods, except OMK 60, are also used in the Swedish national EM program, as previously described (Boye et al., 2019). In total 148 substances were analyzed in one or more samples from this study. However, only 27 of the analyzed substances were permitted for use in Swedish greenhouse production (or 28, if including the thiophanate methyl degradation product, carbendazim) during the study period (SI, Table SI-1) and, therefore, are the focus of this study. Of those 28, six had no registered use in the greenhouses during the study period.

The method used for each of the 28 substances (including the non-detected ones) is reported in the SI (Table SI-1), including limits of detection (LOD), and limits of quantification (LOQ). Most substances had about 2–10 times lower LOD in TIMFIE than grab samples (SI, Table S1) due to the in-field SPE leading to an up-concentration of TIMFIE samples.

All methods used in this study measures the total water concentrations, thus including both dissolved and suspended particle-bound fractions of the substances. It should further be noted that concentrations are dependent on water discharge and, therefore, cannot be used to directly compare total pesticide loading between areas. Such calculations would require flow-proportional sampling, which is much more expensive and technically challenging and was not available for this study. Nevertheless, in terms of toxicity to aquatic organisms, concentrations are the best indicators.

2.3.2. Environmental impact assessment criteria

The potential environmental impact of the pesticide occurrences in the streams was assessed by comparing the measured concentrations with the Predicted No-Effect Concentration (PNEC) calculated from toxicity data included in the most recent European Food Safety Authority (EFSA) conclusion report or the renewal assessment report (RAR; for substances without an EFSA conclusion) for each substance during the registration process (EFSA, 2003–2021). To determine the PNEC values, the lowest value used for calculating the toxicity/exposure ratio (TER) was divided by the EFSA-established assessment factor (AF, normally 10). The toxicity data and references used for the PNEC calculations are listed in the SI (Table S2).

2.3.3. Swedish National Environmental Monitoring Program for pesticides

In order to assess the impact of greenhouse production on pesticide occurrence and potential risk in surface waters, it is necessary to consider the potential contribution from open-field production that surrounds the greenhouses and drains into the same stream. For this purpose, the data from the current study was compared with data from the same time period from two catchments (14 and 8 km^2, located in the same climatic/geographic regions

as the streams from this study) that are included in the Swedish National EM Program for Pesticides, which is conducted in small catchments without greenhouse production. Details and data from that program have been described previously (Boye et al., 2019). Briefly, a sample is taken, from the outlet stream draining a small agricultural catchment, by an automatic ISCO sampler (6712FR) every 90 or 180 min and added to a bottle that is replaced every 7 days (May–November, 90-min sampling interval) or every 14 days (December–April, 180-min sampling interval), respectively. The sample bottles are contained in a refrigerated (4 °C) unit, shipped on ice to the laboratory, and analyzed by the same methods employed for the grab samples from this study.

3. Results and discussion

3.1. Detected substances

Of the 28 substances that were permitted for greenhouse use (including the thiophanate methyl degradation product, carbendazim) and analyzed, 23 were detected in one or more of the grab samples (Table 3) and the same substances were also detected in one or more of the TIMFIE samples, except aclonifen (not analyzed in TIMFIE samples). In addition, fenhexamid, indoxacarb, and pyriproxyfen, which were never detected in grab samples, were detected in at least one TIMFIE sample. Thus, in total 25 substances of the 28 permitted for greenhouse use and analyzed in this study were detected in at least one sample, although only 22 (counting carbendazim as "used" where thiophanate methyl was used) had a registered use in the greenhouses during the study period (Fig. 1). The three substances with no registered use that were never detected in any of the grab or TIMFIE samples were fenpyrazamine, fenpyroximate, and kresoxim-methyl. The other three permitted and analyzed substances with no registered use during the investigation period, aclonifen, indoxacarb, and thiacloprid, were detected in at least one sample.

Table 3
The 23 substances that were detected in grab samples and were permitted for use in greenhouses during the study period, presented with detection frequency (as percent of total number of samples where the substance was analyzed), the maximum concentration measured in one sample, detection frequency above PNEC, and the highest exceedance of PNEC (as quotient between measured concentration and PNEC, when measured concentration > PNEC).

	PNEC (μg/L)	Det. freq.	Max Conc. (μg/L)	Det. freq. > PNEC	Max. Quotient
acetamiprid	0.024	18%	9.4	6%	391
aclonifen[a]	0.5	2%	0.066	0%	
azoxystrobin	0.95	68%	9.2	1%	10
boscalid	12.5	70%	0.76	0%	
carbendazim[b]	0.15	49%	8.9	5%	59
cyprodinil	0.82	37%	0.75	0%	
fludioxonil	0.5	39%	2.9	2%	6
hexythiazox	0.61	12%	0.7	1%	1
imazalil	18.14	7%	1.2	0%	
imidacloprid	0.009	91%	13	44%	1444
mandipropamid	7.6	12%	0.018	0%	
metalaxyl	100	51%	0.64	0%	
paclobutrazol[c]	0.82	1%	0.041	0%	
penconazole	3.2	9%	0.27	0%	
pirimicarb[c]	0.09	34%	3.7	4%	41
propamocarb	630	51%	107	0%	
propiconazole	6.8	43%	1.7	0%	
pymetrozin	2.5	26%	9.2	3%	4
pyraclostrobin	0.3	15%	0.041	0%	
pyrimethanil	30	1%	0.023	0%	
thiacloprid[a]	0.077	19%	0.038	0%	
thiophanate methyl	16.1	7%	6.6	0%	

[a] Substance had no recorded GH use in the study areas but was permitted for GH production in 2017 and 2018.
[b] Carbendazim is a degradation product to thiophanate methyl.
[c] Substance was exclusively permitted for greenhouse production during the study period.

Imidacloprid was the most frequently detected substance in the grab samples (Table 3), in spite of only representing 1.2% of the number of applications in greenhouses in the study areas (Fig. 1). It was also the substance that most frequently exceeded its PNEC value (0.009 μg/L) in both grab and TIMFIE samples (48% of grab and 20% of TIMFIE samples exceeded PNEC) and it exceeded its PNEC value by the largest quotient in both grab and TIMFIE samples; max concentrations were 1444 and 355 times higher than PNEC, respectively. This is likely partially due to the fact that imidacloprid had the second lowest PNEC value of all substances included in this study (pyriproxyfen, detected in one TIMFIE sample, being the only substance included in this study with a lower PNEC than imidacloprid, SI Table S2). It should also be noted that the highest concentration in grab samples for imidacloprid was detected in V7, where TIMFIE samplers were not installed, which may explain the discrepancies in max exceedances between the sampling methods. The detection frequency for imidacloprid in TIMFIE samples was lower (33%) than in grab samples, probably because this substance was one of few that had a higher LOD (0.005 μg/L) in TIMFIE than in grab samples (0.001 μg/L). Indeed, the measured concentrations in grab samples were lower than 0.005 μg/L (the LOD for TIMFIE samples) in 1/3 of the samples where imidacloprid was detected. One possible explanation for the high detection frequency of imidacloprid is that this substance is quite persistent in both water and soil (DT50 = 90 days and 77–82 days, respectively (EFSA, 2014b)), meaning that previous use in greenhouses (prior to the study period) could have contributed although not included in the pesticide application journals. Further, imidacloprid is the most sold insecticide globally (Jeschke et al., 2011) and was the third most sold active insecticide substance on a weight basis in Sweden in 2017 (Kemi, 2017). This substance is frequently detected in the Swedish EM program (Boye et al., 2019), as well as elsewhere (Benton et al., 2016; Mahai et al., 2019; Morrissey et al., 2015; Starner and Goh, 2012; van Dijk, 2010; Vijver et al., 2008; Xiong et al., 2019). Thus, the high detection frequency in this study is not surprising and there are likely other contributing sources than greenhouse use in the catchments. Nevertheless, the detection frequencies in the current study (both total and above PNEC) in locations downstream of greenhouses with recorded imidacloprid use were higher than in study areas without registered greenhouse use, as well as for the EM program in 2017 and 2018 (Fig. 2). Moreover, there were multiple samples in this study that had concentrations of imidacloprid that were higher than the maximum concentration, 0.16 μg/L, found in the EM program during the same period (Fig. 3) and the maximum concentration detected in this study, 13 μg/L, was almost two orders of magnitude higher than that of the EM program (Table 3). Thus, it is highly probable that greenhouse use, during and/or prior to the study period, contributed to the occurrence of this pesticide in the streams.

Aside from imidacloprid, other substances with high detection frequencies (>50%) in grab samples were azoxystrobin, boscalid, metalaxyl, and propamocarb (Table 3, Fig. 2). Acetamiprid was also the substance with the second highest detection frequency above its PNEC value in grab samples (Table 3). Like imidacloprid, azoxystrobin and metalaxyl were not frequently used in the greenhouses upstream of the sampling locations, whereas acetamiprid, boscalid, and propamocarb were among those most frequently used (Fig. 1). All of these substances were also detected at higher concentrations in several grab samples from this study than in the EM program for 2017–2018 (Fig. 3) – some (*e.g.,* acetamiprid) were not detected in any EM samples – further supporting a link between greenhouse use and stream water occurrences. In fact, 18 of the 20 substances that were permitted for greenhouse use, analyzed in both studies (paclobutrazol and pyrimethanil were not analyzed in the EM program), and detected in at least one grab sample in the current study, had a higher maximum concentration in the current study than in the EM program (Fig. 3), with the exceptions being mandipropamid and thiacloprid (Table 3, Fig. 3). Notably, 14 of the 20 substances also had a higher median concentration in samples from the current study than in the EM samples, with boscalid and propamocarb among the remaining six having very similar medians in both studies; this leaves imidacloprid, mandipropamid, metalaxyl, and thiacloprid with a

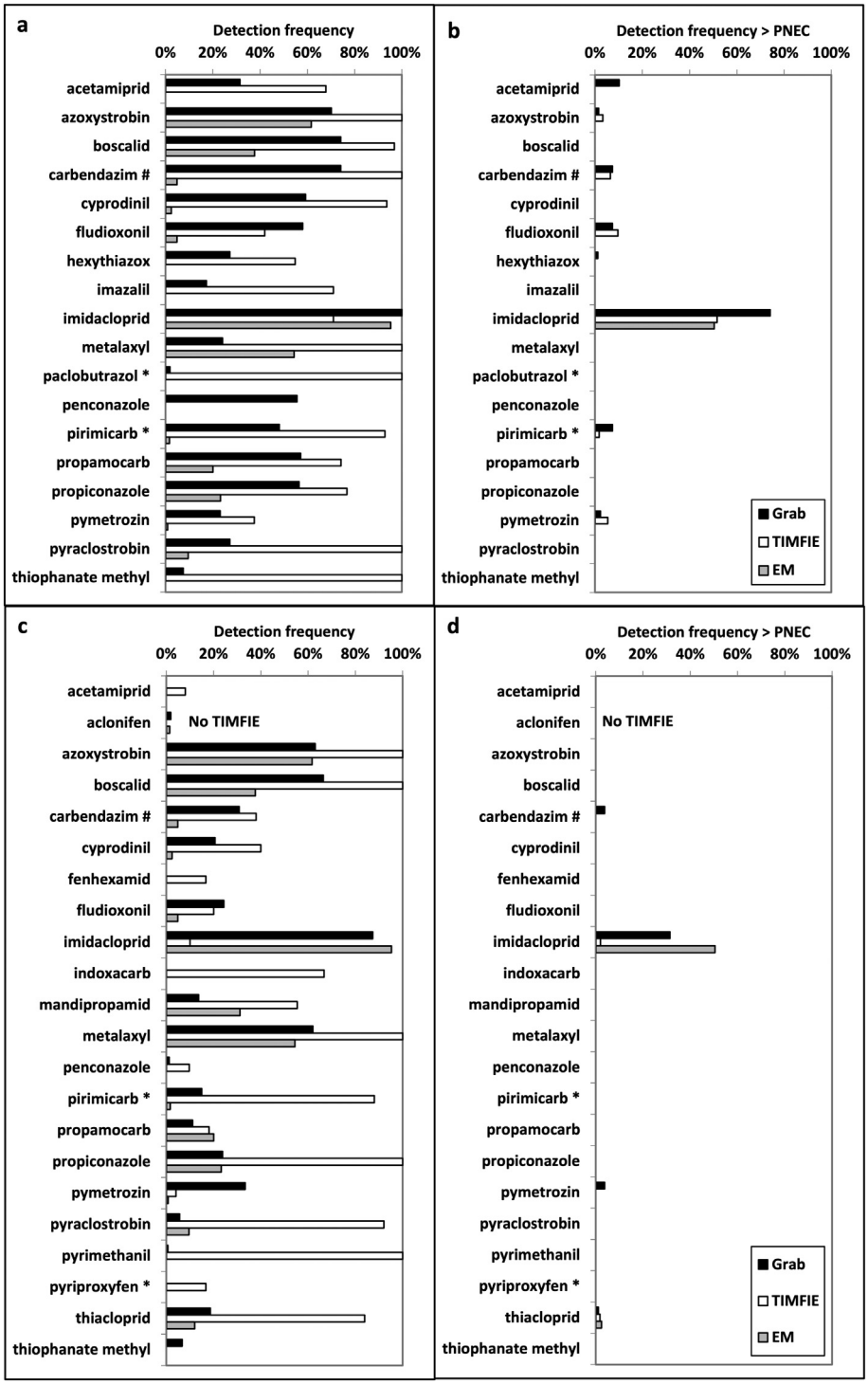

Fig. 2. Detection frequencies for substances with (top) and without (bottom) registered use in the same area. Frequencies of total detections (a) and of detections above PNEC (b) for substances with reported use in greenhouses in the same area where they were detected during the study period. Panels (c) and (d) show the frequencies of detection – total (c) and above PNEC (d) for substances that were permitted for greenhouse use, but had no registered applications in the area(s) where they were detected during the study period. Results from this study (grab samples, black bars and TIMFIE samples, open bars) are shown together with results from the national Swedish EM program of pesticides in surface water in two model catchments (with no greenhouses) from southern Sweden (EM, grey bars) (Boye et al., 2019). # Carbendazim is included as "used" where its parent substance (thiophanate methyl) was used. * Substance was exclusively permitted for greenhouse production during the study period. Note that TIMFIE samples were only analyzed in four of the study areas (O1, O2, OV4, and V5) and that the detection limit for most substances in TIMFIE samples was considerably lower than in grab samples. Thus, direct comparisons between the methods is not possible.

distinctly higher median concentration in EM watersheds, *i.e.* without greenhouse productions (Fig. 3). Thiacloprid had no registered use in the greenhouses in the current study during the study period, and mandipropamid was applied once in one greenhouse (representing 0.1% of the total number of applications in all greenhouses) (Fig. 1). Metalaxyl applications represented 0.6% of all applications (two times each in two greenhouses) (Fig. 1) and had a lower detection frequency, even in the catchments where it was used, than in the EM samples (Fig. 2). This indicates that

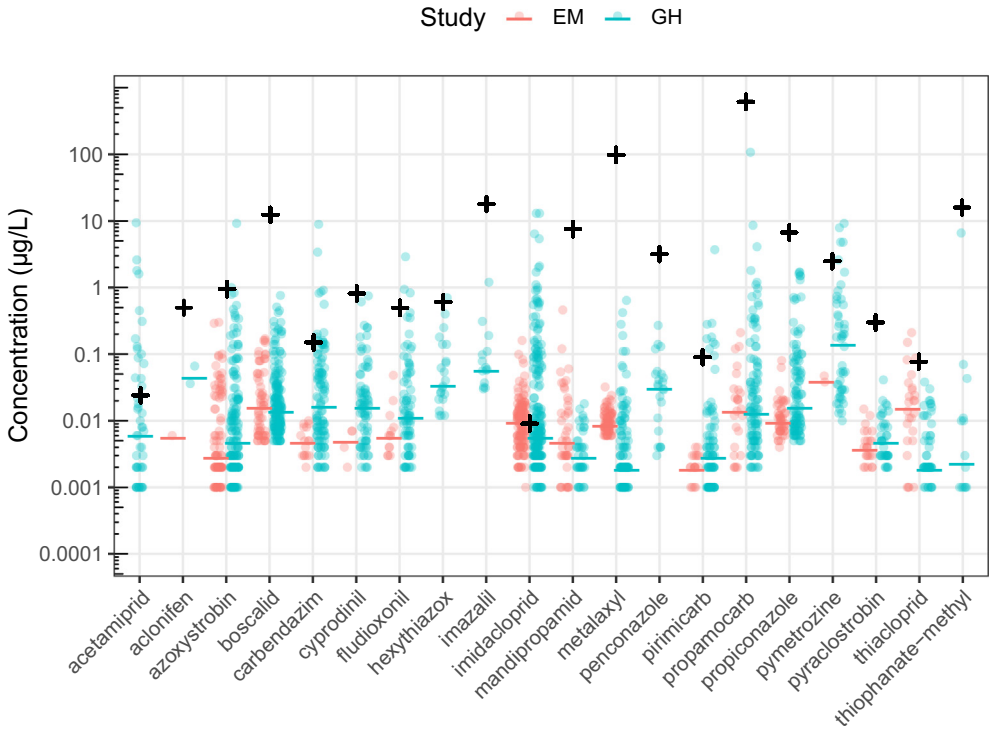

Fig. 3. Concentrations (µg/L, log scale) of substances permitted for use in greenhouses during the study period (2017–2018) and detected in at least one grab sample. Concentrations in grab samples from the current study (*i.e.* in streams with upstream greenhouse production) (blue) compared to concentrations in samples from streams in agricultural catchments without upstream greenhouse production from the Swedish EM program for pesticides during the same period (2017–2018) (red). Median concentrations for each substance within all samples where the substance was detected (non-detects excluded) for each dataset is indicated by horizontal lines in corresponding color and the PNEC value for each substance is indicated by a black plus sign. Note that paclobutrazol and pyrimethanil were not analyzed in the EM program and, hence, are not included in this figure.

thiacloprid, mandipropamid, and metalaxyl occurrences and concentrations in the streams in this study are primarily from other sources than the greenhouses. As for imidacloprid, it is more difficult to decipher the source(s) given its widespread use; in 2017 there were around 20 different products containing imidacloprid registred for use in Sweden, either as a plant protection product (in greenhouses, in forestry, and as seed treatment in agriculture) or as a biocide. Nevertheless, as discussed above there are indications that greenhouses did indeed contribute to its occurrences and, probably more importantly, its concentrations in Swedish streams, although there were other contributing sources. For all of the remaining 17 substances (including boscalid and propamocarb) it appears even more clear that greenhouse use contributed to the number of detections and total concentrations in the streams examined in this study, although the extent of the greenhouse contributions cannot be determined.

The TIMFIE samples had a higher detection frequency than grab samples for most of the analyzed substances (Fig. 2), which primarily should be attributed to the lower LODs for all substances except imidacloprid and propamocarb (higher LOD in TIMFIE than grab samples), as well as indoxacarb and thiophanate methyl (same LOD in TIMFIE and grab samples) (SI, Table S1). Thus, both imidacloprid and propamocarb, exhibiting higher LODs in TIMFIE samples, were detected in less than half of the TIMFIE samples (33% and 40%, respectively). However, azoxystrobin, boscalid, and metalaxyl (*i.e.* with lower LODs in TIMFIE than grab samples) were detected in 100%, 99%, and 100%, respectively, of the TIMFIE samples (Fig. 2). In addition, carbendazim, thiacloprid, pirimicarb, propiconazole, and pyraclostrobin were detected in at least 50% of the TIMFIE samples (Fig. 2). This shows that the LODs are very important for assessing pesticide occurrences. In addition, time-integrated samples (like TIMFIE) may capture a more complete picture of pesticide occurrences and overall average concentrations, than do grab samples (which may hit or miss concentration peaks without capturing the circumstances to determine the causes for concentration variances). However, a direct comparison between the two sampling methods cannot be done in this study because of the differences in analyzed substances, LODs and LOQs, and the fact that TIMFIE samplers were only installed in a subset of the study areas.

3.2. Relevance of greenhouse production system to pesticide occurrences in streams

The number of detected substances and, especially the total number of detections, varied widely between the sampling points, with those downstream of ornamental producing greenhouses generally exhibiting the highest number of detections of used substances (Table 4). The exception

Table 4
Number of detected substances and the total number of detections for substances used in greenhouses from each study area, as well as for substances that were not used in the greenhouses during the study period, but that were permitted for use in greenhouses in Sweden during that time period (note that all except three, paclobutrazol, pirimicarb, and pyriproxyfen, of those substances were also permitted for open-field agriculture and/or other use).

Study area	# of detected, used substances	Total detections of used substances[a]	# of detected, unused, permitted substances	Total detections of unused, permitted substances
O1	3	24	11	98
O2	8[b]	65[b]	5	38
O3	15[b]	221[b]	2	27
OV4	8	159	14[b]	152[b]
V5	7	70	11	115
V6	2	3	9	32
V7	4	64	11[b]	176[b]

[a] Of those used in the greenhouses during the study period.
[b] Carbendazim is counted instead of thiophanate methyl (thiophanate methyl was never detected without carbendazim being detected in the same sample).

to that trend was O1, which had the second fewest number of detections of substances with registered greenhouse use among the catchments, in spite of only having ornamental greenhouse production upstream of the sampling site (Table 4). This catchment was, however, also by far the largest (*i.e.* more water and, hence, dilution) and had only one greenhouse producer (*i.e.* less potential contribution from greenhouse production) (Table 1).

Because all but three (paclobutrazol, pirimicarb, and pyriproxyfen) of the detected substances were also permitted for other uses than greenhouse production during the sampling period, it is not possible to definitively pinpoint the specific contribution of pesticides from greenhouses – either in terms of active or historical use (*e.g.,* in the case of imidacloprid and other long-lived substances). However, the pesticide use journals recorded by the greenhouse producers enabled us to separate out detections of pesticides that had been used from those that had not been used during the study period within the greenhouses in each catchment. Thus, some inferences can be made about greenhouse contributions by comparing number of detections (Table 4) and detection frequencies (Fig. 2) between used and unused substances. To this end, the detections of substances that had no registered use in greenhouses upstream exhibited a different pattern compared to those that had been used (Table 4); O1 and all of the areas with purely vegetable greenhouse production systems (V5, V6, and V7) had more detections of unused substances than used ones. The number of detected, unused substances and the total number of detections of unused substances were also higher in O1, V5, and V7 than in the remaining two catchments with purely ornamental greenhouse production systems (O2 and O3). Although inconclusive, this suggests that ornamental greenhouse production may contribute more to the total number of detected substances and detections than vegetable production systems (considering the above-mentioned potential reasons for why O1 deviated from this pattern). Indeed, the only catchment with both types of greenhouse production systems, OV4, supports this inference by exhibiting similar and relatively high numbers of detections of both used and unused substances (Table 4). OV4 was also the smallest catchment, which further explains the high numbers of detected substances and total detections in this study area. It should be noted, however, that a larger number of substances detected does not necessarily entail higher toxicity or concentrations, but if, as in this case, the findings are consistent with a larger number of pesticides used and application frequency within greenhouses, it does indeed support the specific contribution of greenhouse production to surface water pesticide occurrences. Greater number of detected substances and total occurrences downstream of ornamental facilities is not surprising, given that a larger number of substances were used in ornamental greenhouses (again considering O1 as an exception) and also more frequently compared to the vegetable greenhouses (in this case also valid for O1) (Table 1).

3.3. Greenhouse contributions to overall pesticide occurrences in Swedish stream

Another way to assess the contribution to pesticide occurrences in streams from greenhouses is to compare measured concentrations over time to logged application times in individual catchments (Fig. 4). In this context, it is important to keep in mind the potential lag times from application to substances reaching the stream, depending on substance properties, cleaning of equipments *etc.* that may offset any temporal relationships. Nevertheless, from such a comparison it appears that acetamiprid and pirimicarb use within the ornamental producing greenhouses (O1, O2, and O3) could potentially be linked to elevated concentrations in the respective streams (Fig. 4). Similarly, in the vegetable producing greenhouses propamocarb applications in V5 and azoxystrobin in V6 appear to closely precede elevated concentrations of those substances occurring downstream (Fig. 4). Moreover, substances with frequent and regular applications in greenhouses are consistently detected at the downstream surface water sampling locations, *e.g.,* boscalid (O2, O3), carbendazim (compared to thiophanate methyl use in O3), and propiconazole (O3) (Fig. 4). However, there is no overall consistent pattern that arises between application times and measured concentrations. In addition to the previously discussed possibilities for a disconnect between application times and detects (*e.g.,* transport properties, substance persistence, equipment cleaning), this is very likely due to contribution of pesticides from other sources - mainly open-field agriculture, as suggested by the overall highest concentrations occurring during the agricultural cropping season, May–September. For example, boscalid was detected in 99% of the TIMFIE samples (Fig. 2), even though it was only applied in one of the study areas (O2) where TIMFIE samplers were installed (Fig. 4), clearly indicating other sources than greenhouse production must have contributed to the occurrence of this substance. The lack of consistent patterns between applications and detections may also indicate differences in greenhouse facilities, in terms of, for example, the water handling, pesticide application approach (spraying *vs.* watering), and/or the extent of covered/sealed surfaces – *i.e.* how closed and leak-proof the system is. As an example, OV4 hosts 5 greenhouses (2 ornamental and 3 vegetable producing facilities), represents the smallest of the catchments included in this study (*i.e.* least possibility for dilution and/or contribution from alternative sources), and had the third largest number of pesticide applications during the study period (Table 1) – yet, the most applied substance, imazalil, was only detected in 7% of the samples (data not shown) and there is no discernible correlations between application times and measured concentrations for any of the used substances (Fig. 4). On the other hand, O2 exhibited increased concentrations of substances recently applied in the greenhouses within that area (*e.g.,* acetamiprid detection in July 2017 and pirimicarb in July 2017 and September 2018). It is, however, not possible to draw any clear conclusions from the comparisons of application times and surface water concentrations in individual catchments due to the multiple complicating factors, *e.g.,* catchment sizes, number of greenhouse facilities, other permitted pesticide uses, potential recent use of pesticides in greenhouses prior to the study period (*i.e.* not recorded within the application journals), substance persistence, as well as any cleaning or other non-recorded and/or irregular activities that may have resulted in point-source or otherwise unusually high pesticide leaching from the greenhouses – in addition to any other sources within the catchment.

Finally, in an attempt to further evaluate greenhouse contributions to surface water occurrences and concentrations of pesticides in Swedish streams, we compared pesticide concentrations in up- and downstream locations relative to the greenhouses in three of the catchments in this study (O1, O2, and V6) (Fig. 5). All of the differences between up- and downstream concentrations were very small (<0.053 μg/L diffference) or non-existent and several of the substances that exhibited differences had no registered use in the greenhouses in the same catchment during the study period (Fig. 5). For example, imidacloprid had no registered use in any of the three catchments with upstream/downstream sampling during the study period and the differences between up- and downstream concentrations varied from positive to negative between time points with no consistent pattern. Nevertheless, these comparisons provide some support for the indications noted previously; that greenhouses do contribute to pesticide occurrences in Swedish streams, especially in terms of substances that are frequently used in the greenhouses (Fig. 1), such as acetamiprid, boscalid, thiophanate methyl (as indicated by detections of the degradation product, carbendazim), and pirimicarb; all four of those substances were detected in higher concentrations in the downstream compared to the upstream sampling points in the areas where they were used (Fig. 5). These substances also had higher detection frequencies in areas where they had been used in greenhouses in this study compared to the EM results and lower detection frequencies, except for boscalid, in areas where they had not been used in the greenhouses (Fig. 2). Notably, propamocarb and propiconazole also exhibited higher detection frequencies and higher (propiconazole) or similar (propamocarb) median concentrations in grab samples from this study, compared to the EM program, downstream of greenhouses where they were used, but lower (propamocarb) or similar (propiconazole) detection frequencies where they were not used (Fig. 2). However, the TIMFIE data for propiconazole (Fig. 2) and the upstream/downstream comparisons for propamocarb (Fig. 5) do not align with that observation. It should be noted, though, that the TIMFIE samplers were only installed in two of the same catchments (O1 and O2) where the up-

and downstream grab sampling took place and each of the two substances were only used in one of the catchments with up- and downstream sampling points (propamocarb in V6, and propiconazole in O1), making cross-comparisons less reliable. Moreover, both propamocarb and propiconazole were more frequently detected in the downstream locations of greenhouses where they had been used (Fig. 2) and typically at higher concentrations (Fig. 4) than where there was no recorded use. Thus, it still seems likely that greenhouses contribute to the occurrence of these two substances.

4. Conclusions

This study aimed to elucidate the potential contributions of greenhouse production systems to pesticide occurrences and toxicity in Swedish

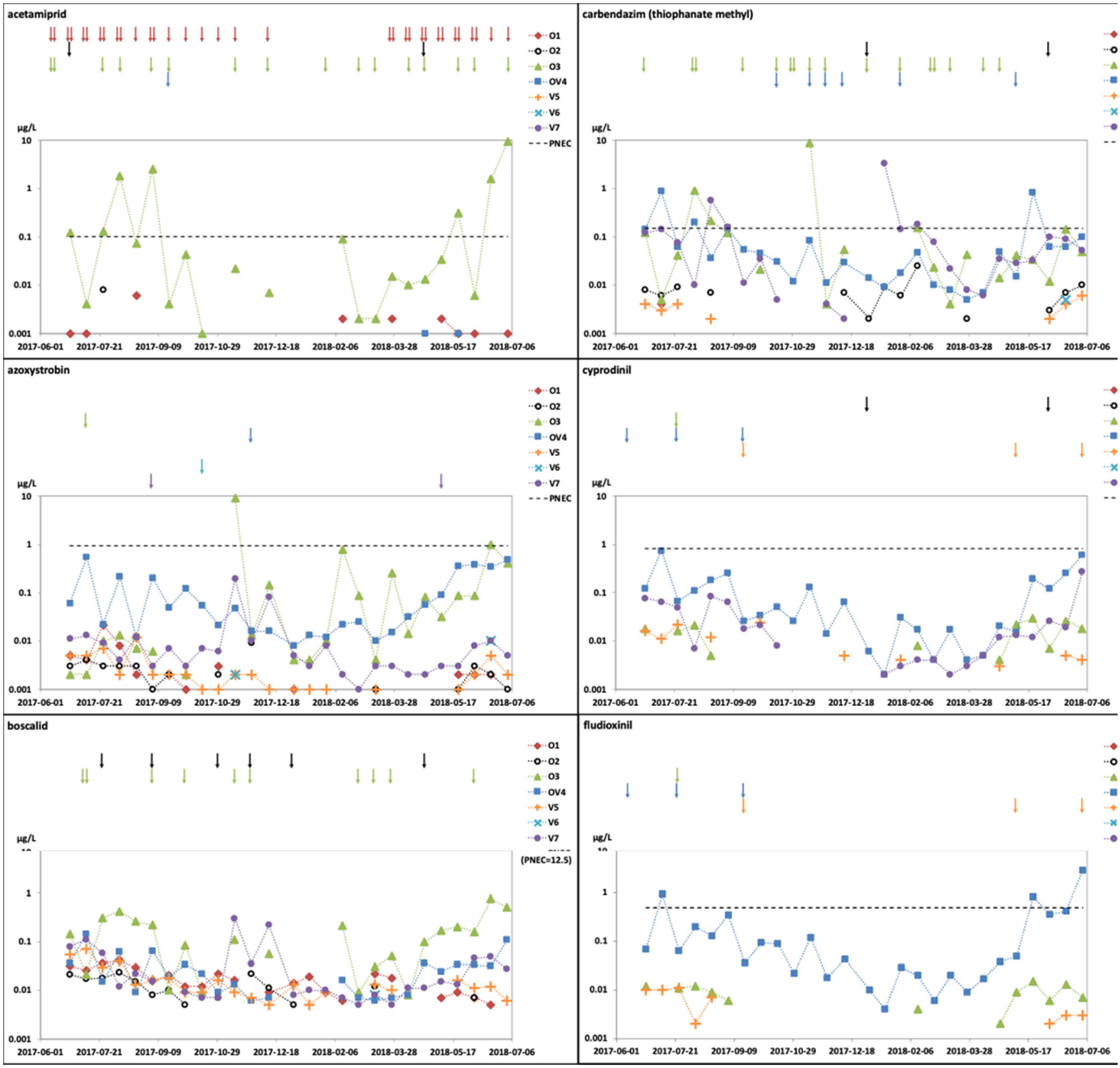

Fig. 4. Concentrations (µg/L, log-scale) of the 12 pesticides that were used in two or more of the study areas and detected in more than 50% of the grab samples and/or detected above its PNEC value in at least one sample within the same study area(s) where they were used. O1 = red diamonds, O2 = black open circles, O3 = green triangles, OV4 = blue squares, V5 = orange pluses, V6 = turquoise x:s, V7 = purple filled circles. The dashed black lines indicate the PNEC values for each substance, except for boscalid (PNEC = 12.5) and propamocarb (PNEC = 630) that had PNECs well above the measured concentrations). Arrows indicate applications in greenhouse(s) within the corresponding (by row and color) study area in the 14-day period preceding each sampling occasion. Two or three arrows close together indicate several applications were made within the same area (same or different greenhouses) within the 14-day period preceding the sampling occasion. Pirimicarb is the only of these 12 substances that was exclusively permitted for greenhouse production during the study period.

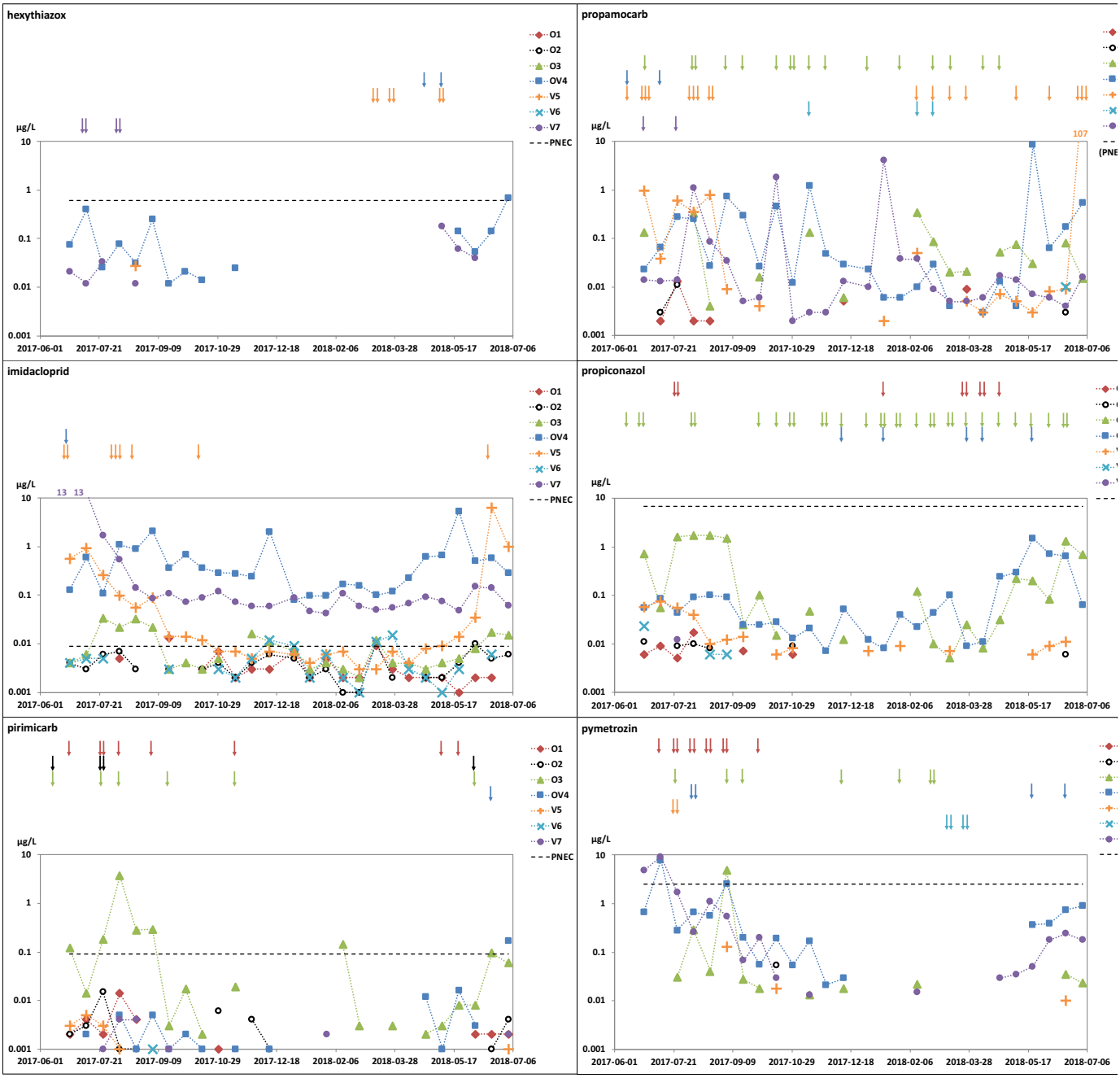

Fig. 4 (*continued*).

streams. While it is not possible to quantify the relative contribution from various sources of pesticide occurrences in streams in catchments with multiple potential pesticide application purposes (including open-field agriculture, greenhouses, turf grass, outdoor nurseries, biocide use *etc.*), we used several approaches to assess the likelihood of greenhouses contributing to pesticide occurrences in seven catchments with at least one greenhouse producer upstream of the sampling location in southern Sweden. Our results confirm the findings of previous studies (Kreuger et al., 2010; Leistra et al., 1984; Roseth and Haarstad, 2010); greenhouses do contribute to pesticide occurrences in surface waters. Most of the substances that were approved for greenhouse use in Sweden during the study period were detected in a higher percentage of samples and/or at higher concentrations in this study (which focused on catchments with greenhouse production) compared to the Swedish EM program (with no greenhouse production upstream of the sampling locations). This was particularly true for substances that were applied frequently within the greenhouses. Ornamental greenhouse production generally appeared to contribute more often to the occurrence of pesticides (regarding detection frequencies and numbers of detected substances, but not necessarily concentrations), likely due to the higher frequency of pesticide applications and number of substances used, in addition to the year-round production.

In terms of ecotoxicity, the substance detected in concentrations of greatest concern (both in terms of maximum exceedance and number of detected exceedances of its PNEC) within this study was imidacloprid. Although the evidence is convoluted by the wide-spread application of this substance across multiple types of uses, as well as its relatively long persistence in soil and water, our data suggests that greenhouses indeed were important contributing sources to the occurrences and, more importantly, the total concentrations (frequently above its PNEC) of this substance in streams within this study. It should be noted that, as of December 12,020,

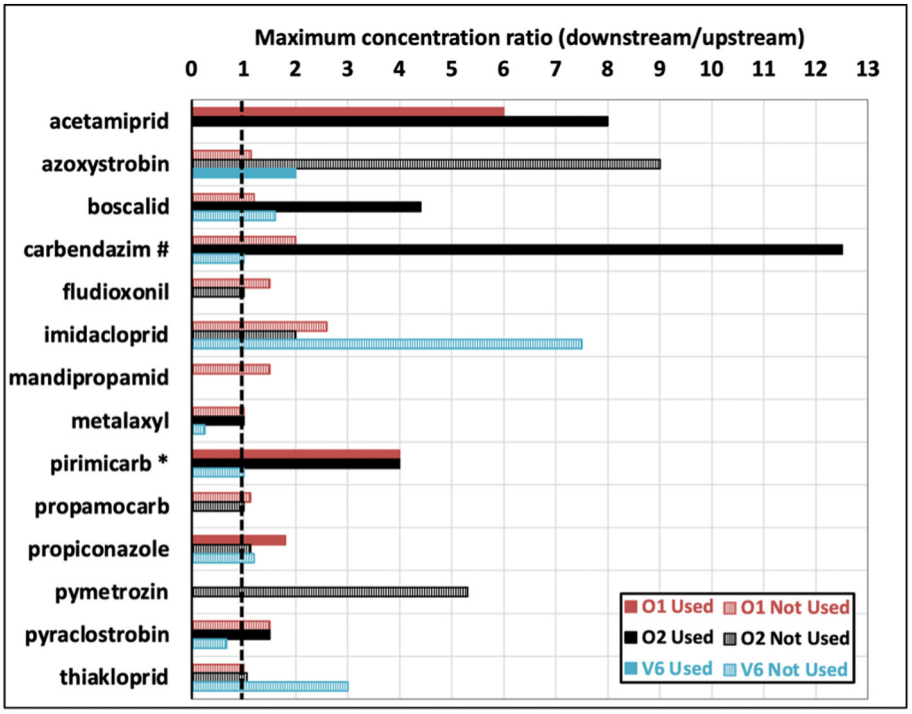

Fig. 5. Largest ratios of concentrations measured downstream relative to upstream of greenhouses in O1, O2, and V6 areas at any time point during the study period. Only substances detected in at least one of the study areas are included. Striped bars indicate the substance was not used in the greenhouses within that area during the study period. # carbendazim was not used in any of the study areas, but it is a degradation product to thiophanate methyl, which was used in O2. Thiophanate methyl was not detected in any of the samples (up- or downstream) in any of these three study areas. * pirimicarb was exclusively permitted for greenhouse production during the study period.

imidacloprid is no longer permitted for plant protection use (including open-field and greenhouse productions) in the EU; however, it is still permitted as a biocide.

It is notoriously difficult to pinpoint sources for pesticide occurrences in surface waters due to the (normally) multiple allowed uses, paucity of data regarding all actual application dosages, uses, and timing in the upstream watershed, as well as the immense complexity that arises from substance/soil/hydrological properties, practices of the farms/greenhouses/other facilities that use (or historically used) a specific pesticide *etc.* Nevertheless, the data presented here indicates that pesticide leaching from greenhouses is not limited to soil, air, and/or groundwater contamination and that greenhouse worker and/or consumer exposure is not the only concern related to pesticide use in these systems. Indeed, our study suggests that greenhouses do contribute to surface water occurrence and potential toxicity of pesticides, although the extent is likely dependent on, for example, the type of greenhouse production system (*e.g.,* ornamentals *vs.* vegetables), engineering and infrastructure, water handling, and pesticide application practices.

Hence, leaching models and other risk assessment tools should be taking into account the potential contributions from greenhouses to surface water concentrations of pesticides. Moreover, efforts should be put towards understanding the routes of exposure from greenhouses to surface water within a holistic context of the watershed (*e.g.,* including indirect transport *via* air and groundwater, considering the specific infrastructure, practices *etc.* for different greenhouses, and how those factors combine with specific substance properties to increase or decrease risks for leaching into surface waters), in order to help develop efficient mitigation strategies specific to greenhouse production and pesticide use.

CRediT authorship contribution statement

Kristin Boye: Conceptualization, Formal analysis, Visualization, Writing – original draft. **Gustaf Boström:** Data curation, Formal analysis, Investigation, Visualization, Writing – review & editing. **Ove Jonsson:** Investigation, Methodology, Validation, Writing – review & editing. **Mikaela Gönczi:** Project administration, Funding acquisition, Writing – review & editing. **Klara Löfkvist:** Investigation, Writing – review & editing. **Jenny Kreuger:** Conceptualization, Data curation, Funding acquisition, Project administration, Supervision, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary information

Supporting information for this publication is available: Appendix A - analyses and toxicology information for substances analyzed and permitted for use in greenhouses in Sweden during the study period; Appendix B - complete dataset of concentrations for all 148 substances (including those not permitted for greenhouse use during the study period) analyzed in grab samples from this study. Supplementary data to this article can be found online at doi:https://doi.org/10.1016/j.scitotenv.2021.152215

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