ChEmiTecs

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IVL Swedish Environmental Research Institute

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Assessing the relative importance of emissions from articles of selected organic substances

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ChEmiTecsis a research program funded by the Swedish EPA. The program's goal is to improve the understanding of emissions of organic substances from articles and to clarify and determine the magnitude of this problem. The program aims to support development of Swedish and EU management programs to minimise risks from harmful substances. The program started in December 2007 and proceeded until December 2013. Participating organisations and organisation representatives are:

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Summary

This report presents the results from a nation-wide substance flow analysis (SFA) of diuron, valid for Sweden. The report provides a complement to a previous study in the ChEmiTecs prorgramme published by Westerdahl et al. (2012). The results from the SFA of diuron are summarised in Table 1.

Table 1. Emissions and flows of diuron in Sweden.

Source	Name flow	Size flow (kg/year)	Released to
Households and industry	Diffusion to air	1.3 kg	Indoor air
Households and industry	Influent wastewater	60 (5-175)	M-WWTP
WWTP	Effluent wastewater	60 (<20-140)	Surface water
WWTP	Sewage sludge	<0.6 – 8 kg	Soil (filling material and agricultural soil)
WWTP	Sewage sludge	<0.5 – 3 kg	Landfill
WWTP	Sewage sludge	<0.005 – 0.03 kg	Incineration
WWTP	Sewage sludge	<0.5 – 3 kg	Unknown
Air pollution	Atmospheric deposition	0-2 kg	Inland surface water
Air pollution	Atmospheric deposition	0-30 kg	Soil

A substance flow diagram for diuron in Sweden is presented in the Figure 1. The values are based on most recent data found and are presented in kg diuron per year.

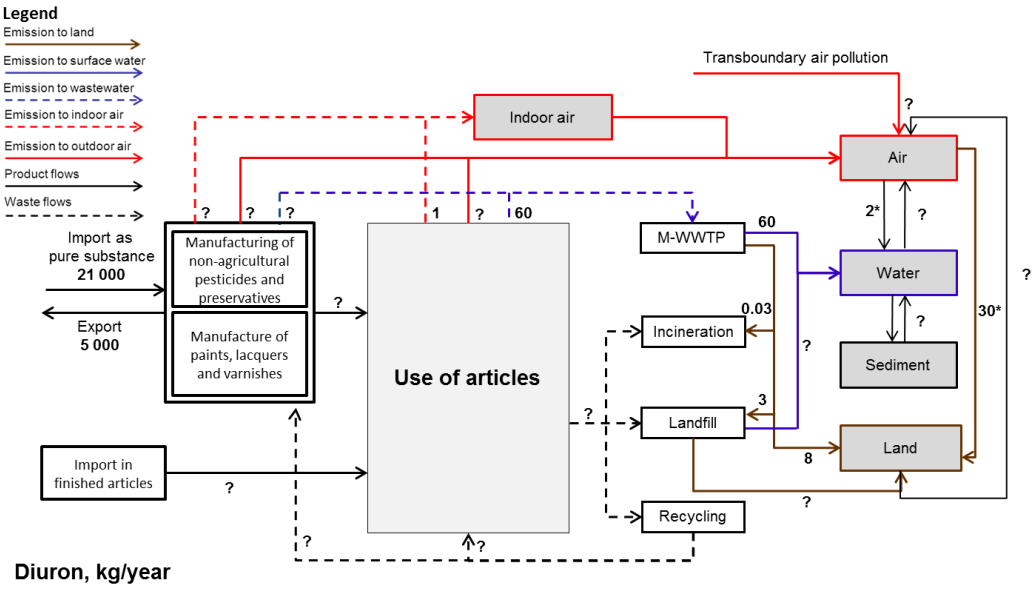


Figure 1. Substance flow diagram for diuron in Sweden (kg diuron/year). * marks flows where concentrations were below the detection limit.

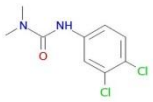
The SFA shows that approximately 20 000 kg diuron is imported to Sweden as a pure substance on an annual basis. The inflow of diuron in imported articles has not been possible to investigate in this study. Emissions during service life of paint for inner walls have been estimated, both during painting and from dry paint. Approximately 60 kg diuron is emitted to wastewater during painting while approximately 1 kg is emitted to indoor air from dry paint each year. It should be noted that the emission estimate to wastewater is a rough estimate. The major known emission sources of diuron to the environment are the M-WWTP.

1 Background information

1.1 Physical and chemical properties

Diuron is a herbicide belonging to the substituted urea herbicides. Diuron is the common name for 3-(3,4-dichloro phenyl)-1,1-dimethyl urea. It is formulated as a powder and as a liquid suspension (US EPA, 2013). The chemical structure and the physical and chemical properties of diuron are shown in Table 2.

Table 2. Chemical structure and physical and chemical properties of diuron (EU-ECB, 2000)

	Diuron
Structure	
CAS no.	330-54-1
Molecular formula	$C_9H_{10}Cl_2N_2O$
Molecular weight (g/mol)	233.10
Density (g/cm^3)	1.48
Melting point (°C)	157
Boiling point (°C)	180-190
Water solubility (mg/l)	35 at 20 °C
Vapour pressure (Pa)	6.90×10^{-8} mm Hg at 25 °C

1.2 Production and use

According to the ESIS, European chemical Substances Information System, diuron is a high production volume chemical (ESIS, 2013). Globally, diuron is used as herbicide to control a wide variety of annual and perennial broadleaf and grassy weeds, as well as mosses. It is also used on many other agricultural crops such as fruit, cotton, sugar cane, alfalfa, and wheat, as well as for non-crop areas (US EPA, 2013).

In 2008, diuron was included/approved as an active substance in Annex I to the EU directive concerning the placing of plant protection products on the market (2008/91/EC[1] amending Council Directive 91/414/EEC[2]). According to the Swedish pesticide register, diuron has been used as an active substance in totally 18 herbicide preparations until year 1992. Today, there are however no new approved herbicidal preparations containing diuron in use in Sweden (KemI-stat, 2013; Pesticides Register, 2013).

According to the Commission Regulation (EC) No 1048/2005 to the Biocidal Products Directive (Directive 98/8/EC[3]), diuron may also be used as active substance for film preservatives (product group 7) and masonry preservatives (product group 10). Since 2009, diuron is no longer approved as an active substance in in-can preservatives (product group 6) and since 2008, use of diuron in antifouling products is neither approved (product group 21) in the EU.

The total use of diuron in chemical preparations in Sweden was approximately 20 tonnes in 2011, see Figure 2. During the time period 1999-2011, the use of diuron has fluctuated

[1] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:262:0031:0033:EN:PDF
[2] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:1991:230:0001:0032:EN:PDF
[3] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:1998:123:0001:0063:EN:PDF

between 4 and 58 tonnes per year, with the highest value 2008. It should be noted that diuron in imported finished articles are not included in these figures. The total net inflow of diuron to the Swedish society could therefore be higher than the net inflows presented above.

Looking at the number of preparations containing diuron, there has been more than a twentyfold increase since the beginning of the studied time period, from six preparations in 1992 to 129 preparations in 2011 (Figure 2).

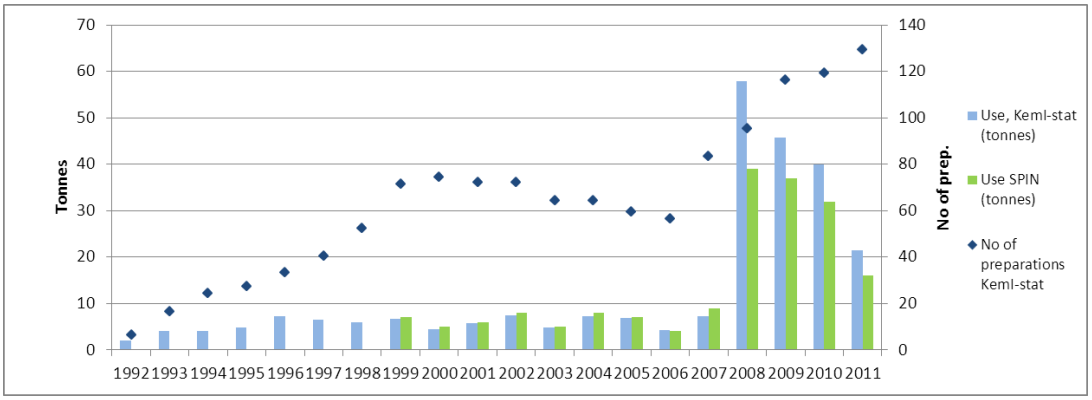


Figure 2. Use of diuron in preparations in Sweden between 1992 and 2011 (SPIN, 2013; KemI-stat, 2013).

Table 3 shows the use of diuron in Sweden divided into different use categories (SPIN, 2013). The major use of diuron in Sweden is as non-agricultural pesticides and preservatives.

Table 3. Use of diuron in Sweden (2011) per national use category (SPIN, 2013).

Code	Use category	No of preparations	Use (tonnes)
39	Non-agricultural pesticides and preservatives	7	10
59	Paints, lacquers and varnishes	89	5.0

1.3 Environmental fate and environmental levels

If released to air, a vapour pressure of 6.90×10^{-8} mm Hg at 25 °C indicates diuron will exist solely in the particulate phase in the atmosphere. The particulate-phase diuron will further be removed from the atmosphere by wet or dry deposition (U.S. National Library of Medicine, 2013).

If released to soil, diuron is expected to have moderate to low mobility based upon measured Koc values ranging from 224 to 879. Volatilization from moist soil surfaces is not expected to be an important fate process. Diuron is not expected to volatilize from dry soil surfaces based upon its vapour pressure (U.S. National Library of Medicine, 2013).

If released into water, diuron is expected to adsorb to suspended solids and sediment and the volatilization from water surfaces is not expected to be an important fate process (U.S. National Library of Medicine, 2013).

Environmental levels of diuron found in the literature are shown in Table 4. Some of these data were used for emission estimates of diuron.

Table 4. Concentrations of diuron in different environmental matrices in Sweden.

Matrix	Concentration Average (Min-Max)	No. of samples	Reference
Ambient air			
Air, background location	<0.5 pg/m^3	3	SEPA, 2013a
Atmospheric deposition			
Atmospheric deposition, background location	<0.07 µg/(m^2 year)	3	SEPA, 2013a
Biota			
Starling	<2 ng/g ww	19	Odsjö *et al*., 2008
Earthworm	<2 ng/g ww	12	Lind, 2011
Water			
Surface water	0.022 (<0.01-0.05) µg/L	125	Sternbeck and Österås, 2010
Sediment			
Sediment	0.34 (<0.05-1.3) ng/g dw	97	Kaj et al., 2010
Sediment	0.038 (<0.002-0.14) mg/kg dw	36	SEPA, 2013b
Soil			
Soil	0.015 (<0.01-0.015) mg/kg dw	6	Törneman and Johansson, 2009
WWTP			
Influent	0.02 (<0.01-0.02) µg/L	2	Sternbeck and Österås, 2010
Effluent	0.034 (<0.01-0.08) µg/L	8	Sternbeck and Österås, 2010
Sludge, M-WWTP	0.063 (<0.01-0.063) mg/kg dw	4	Törneman and Johansson, 2009
Sludge, M-WWTP	<0.01 mg/kg dw	6	Sternbeck and Österås, 2010
Sludge, industrial WWTP	284 mg/kg dw	1	Törneman and Johansson, 2009
Stormwater			
Stormwater	0.22 (<0.01-1.2) µg/L	31	Sternbeck and Österås, 2010

2 Sources of emissions of diuron

2.1 Manufacture

Diuron is not produced in Sweden. Emissions from the production of diuron are therefore not discussed further. According to SPIN (2013) and KEMI (2013) diuron is used within Swedish industry, why emissions during processing of paints, lacquers and varnishes and non-agricultural pesticides and preservatives may occur.

2.2 Service life of consumer articles and materials

No data were found on concentrations of diuron in air and dust from indoor environments in Sweden. Emissions of diuron from indoor paint have been estimated within Project 4 of the ChEmiTecs programme (Molander *et al.,* 2012). The emission estimates were based on data from the Swedish Chemicals Agency's Product registry (KemI-stat, 2013), where the content of chemical products are listed. Figure 3 presents the use of diuron in water based paint for indoor use during the period 2002-2010. The average content of diuron during this time period was 0.002% in wet paint and 0.006% in dry paint.

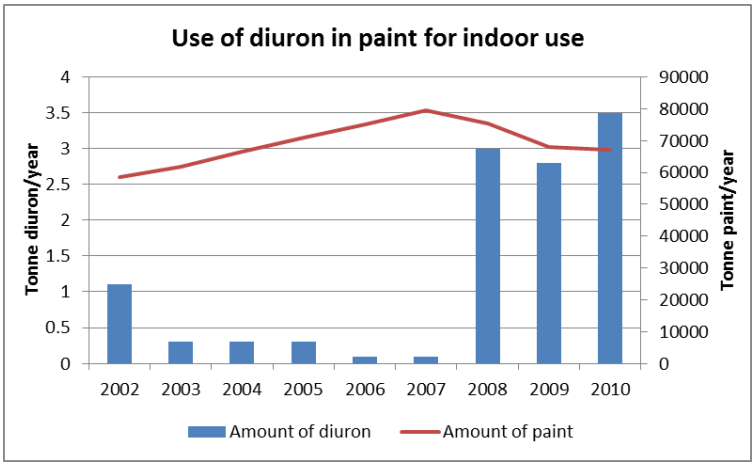


Figure 3. Content of diuron in water based paint for indoor use in Sweden from 2002 to 2010 (KemI-stat, 2013).

Emissions of diuron from painted inner walls (dry paint) were estimated using the emission model by Holmgren *et al.* (2012). Each year, approximately 1.3 kg diuron is emitted to air from dry paint on inner walls in Sweden (Molander *et al.,* 2012). Emissions also take place during painting. Assuming that 5% of the paint is emitted to waste water during cleaning of painting equipment, 60 (5-175) kg diuron is emitted to wastewater each year based on the amounts of paint presented in Figure 3.

2.3 Municipal wastewater treatment plants

Diuron has been measured in influent, effluent and sludge from M-WWTPs in Sweden (Sternbeck & Österås, 2010; Törneman & Johansson, 2009), see Table 4. The average concentration of diuron in effluent water from Swedish M-WWTPs was 0.034 µg/L, with a range of <0.01 to 0.08 µg/L. The yearly outflow of diuron in sewage effluent from Swedish M-WWTPs was estimated to be 60 (<20-140) kg. The methodology for estimation of flows in M-WWTPs is described in Westerdahl *et al.* (2012).

As mentioned above, diuron has also been measured in sewage sludge in an average concentration of 0.063 mg/kg dw (<0.01-0.063 mg/kg dw). Following the approach given in Westerdahl *et al.* (2012), the total amounts released via sludge was estimated to be <2-13 kg distributed according to:

- Filling material: <0.6 – 4 kg
- Agricultural soil: <0.6 – 4 kg
- Landfilling: <0.5 – 3 kg
- Incineration: <0.005 – 0.03 kg

- Other disposal: <0.5 – 3 kg

2.4 Air transport and atmospheric deposition

Diuron is measured in atmospheric deposition at the background station Råö at the Swedish west coast. The yearly deposition, based on the last three years of measurements, was below the detection limit for the analysis (<0.07 μg/m^2 and year). Assuming the same value for the whole country, the annual diuron deposition on inland surface water and on land could be estimated to 0-2 kg respectively 0-30 kg.

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