



Antibiotics residues in pig slurry and manure and its environmental contamination potential. A meta-analysis

Larissa Frey¹ · Benjawan Tanunchai¹ · Bruno Glaser¹

Accepted: 31 January 2022 / Published online: 13 April 2022
© The Author(s) 2022

Abstract

Livestock excrements are used as natural fertilizer, in the form of manure or slurry, to provide nutrients and organic matter to arable soils. However, it is potentially contaminated with antibiotics residues, used in livestock farming to prevent diseases (and thus animal losses), as well as to increase animals' body weight. This poses a contamination risk to surrounding environments. Therefore, we quantitatively evaluated data from 57 peer-reviewed articles published over the past 20 years to generate an overview of antibiotics residues in manure, slurry, soils, plants, and water. Our results revealed that pig manure and slurry, mostly from European countries, contained fluoroquinolones, sulfonamides, and tetracyclines. Furthermore, antibiotics used in animal husbandry are found to contaminate surrounding environments, exceeding the proposed EU threshold value for maximal environmental contamination by veterinary antibiotics in soil in many countries. In soil samples, the highest contents of tetracyclines and sulfonamides were detected in the UK ($41 \pm 18 \mu\text{g/kg}$ and $300 \pm 10 \mu\text{g/kg}$, respectively) and in Austria ($370 \mu\text{g/kg}$) for fluoroquinolones. In water environments, antibiotics contents were detected in low amounts in most countries ($< 2 \mu\text{g/L}$), with the highest content of fluoroquinolones and tetracyclines detected in the USA ($3 \pm 0.7 \mu\text{g/L}$ and $1.3 \pm 0.6 \mu\text{g/L}$, respectively) and of sulfonamides in the USA and Germany ($0.3 \pm 0.8 \mu\text{g/L}$ and $0.2 \pm 2 \mu\text{g/L}$, respectively). In plants, an accumulation of tetracyclines and sulfonamides was found in China, Germany, and Spain ($> 50 \mu\text{g/kg}$). However, no significant specificity of these antibiotics residues to country or continent could be observed. It is urgent that the use of veterinary antibiotics be significantly reduced in order to diminish their residues in slurry, and thus their contamination potential to the surrounding environments.

Keywords Veterinary drug · Environmental contamination · Livestock production

1 Introduction

To maintain proper hygienic conditions of livestock in agriculture, excrements of animals are removed from their housing. The undigested remains, urine, and faeces can subsequently be used as natural fertilizer on arable land in the form of slurry or manure, respectively (Bohrer et al. 2019; Marszałek et al. 2019). To prevent diseases and animal loss during breeding, veterinary antibiotics are frequently used therapeutically in livestock agriculture in many countries (Bohrer et al. 2019; Marszałek et al. 2019). Antibiotics can

also be applied sub-therapeutically to increase the animals' body weight or to improve the efficiency of food intake (Bohrer et al. 2019; Chee-Sanford et al. 2009; Marszałek et al. 2019; Rasschaert et al. 2020). However, due to the chemical structure and bio-availability of the antibiotics, the prescribed drugs may not be completely metabolized and thus absorbed by the animals. The remaining parent compounds and their metabolites of up to 60–90 % will be excreted in faeces and urine (Bohrer et al. 2019; Chee-Sanford et al. 2009; Marszałek et al. 2019). As these components can affect the surrounding environment, its storage and application may create a contamination risk to water, plant, and soil environments. Moreover, with the use of antibiotics, antibiotics-resistant bacteria can develop and consequently be found in the gastrointestinal tract of the animals (Rasschaert et al. 2020; Xia et al. 2019), with a subsequent threat of transfer to humans. Therefore, not only the antibiotics residues but also the resulting resistant bacteria may spread and accumulate in other

✉ Bruno Glaser
Bruno.glaser@landw.uni-halle.de

¹ Soil Biogeochemistry, Institute of Agricultural and Nutritional Sciences, Martin Luther University Halle-Wittenberg, von-Seckendorff-Platz 3, 06120 Halle(Saale), Germany

ecological niches, such as groundwater, surface water and water courses, and soils and crops, depending on the properties of the antibiotics (Bohrer et al. 2019; Kivits et al. 2018; Rasschaert et al. 2020). The input of those compounds into the soil environment may affect its microorganisms, which are responsible for nutrient cycles and pollutant degradation (Marszałek et al. 2019). This contamination may lead to the accumulation of those compounds in plant tissue of crops growing on fields fertilized with slurry or through irrigation with contaminated water (Kivits et al. 2018; Marszałek et al. 2019). Furthermore, this might have an adverse effect on human and animal health as it can contribute to possible development of allergies and antibiotic resistances (Marszałek et al. 2019). Furthermore, antibiotics may have negative effects to ecosystem services such as biodiversity, which is beyond the scope of our study.

According to the One Health Concept, all ecological niches are connected (Destoumieux-Garzón et al. 2018). Residues of raw manure from animals treated with antibiotics may reach livestock or humans when it is being spread on arable land. Due to antibiotic-resistant bacteria, some diseases may become difficult to treat for both animals and humans (Rasschaert et al. 2020). Nevertheless, no threshold values exist to limit the amount of drug residues reaching soil or water bodies in European countries. However, the EU directive CVMP/VICH/592/98-FINAL from 2000 (Committee for Medicinal Products for Veterinary Use 2016) proposes the predicted environmental concentration (PEC) of veterinary medicinal products to be less than 100 µg/kg in soil, and less than 1 µg/L in groundwater. The maximum amount of veterinary drug residues in livestock food products by EU regulation EG 37/2010 (The European Commission 2010) states that no animal tissue may contain more than 100–600 µg/kg. However, there are no threshold values for vegetable food products growing on agricultural land treated with contaminated manure or slurry.

Therefore, knowledge on the quantity of antibiotics contained in manure and slurry as well as their contamination potential in different environments such as soil and water bodies will increase our understanding on sources and magnitude of environmental contamination with antibiotics. Our research focuses on the most frequently used antibiotics in livestock agriculture: tetracyclines, sulfonamides, and fluoroquinolones (Bohrer et al. 2019) in different countries and environmental compartments. Therefore, we aimed at generating a systematic and quantitative overview of veterinary antibiotics residues found in livestock excrements and the environment including soil and water. We will test the following hypotheses:

- i) Residues of different groups of antibiotics can be found in manure and slurry from agricultural livestock.
- ii) Antibiotics residues can be found in the environment including soil, water, and plants directly adjacent to agricultural land fertilized with manure or slurry.
- iii) Type of antibiotics residues is country- and / or continent-specific.

2 Material and methods

2.1 Source of data

The peer-reviewed published articles and book chapters used in this study were obtained from ISI Web of Science. Key words used for the search were a) “pig slurry” and “antibiotic” and b) “pig manure” and “antibiotic.” The time frame was set for the years between 2000 and 2020. A total of 452 entries matching these search properties were found at ISI Web of Science. Subsequently, studies used for this meta-analysis were selected by the following criteria: i) the literature contained the information on the amount of antibiotics detected in manure and slurry given in µg/kg or in any unit which could be converted to µg/kg; ii) the obtained antibiotics contents are based on field data and were not generated artificially; and iii) the obtained contents were available as means or could be calculated as such. Finally, 12 articles containing the amount of antibiotics in pig slurry, 21 articles containing the amount of antibiotics in pig manure, and 24 articles containing the amount of antibiotics in soil and water environments, and plants were evaluated in this study.

2.2 Data categorization

In this study, antibiotics detected in pig slurry and manure as well as soil, water, and plants were grouped into four groups, including fluoroquinolones, sulfonamides, tetracyclines and all other groups of antibiotics.

2.3 Statistical analysis

The statistical analysis was conducted using RStudio (version 1.4.1106). The boxplots were generated using the package “ggplot2” (Wickham 2009). The statistic test used in this study was performed using the package “car” (Fox and Weisberg 2019). Data were checked for normality and equality of variances using Shapiro-Wilk test and Levene’s test, respectively. Since the data are not normal-distributed, a non-parametric Kruskal-Wallis test was used for a comparison of medians between sets of samples. Standard errors were calculated for the median values for each country and antibiotics group.

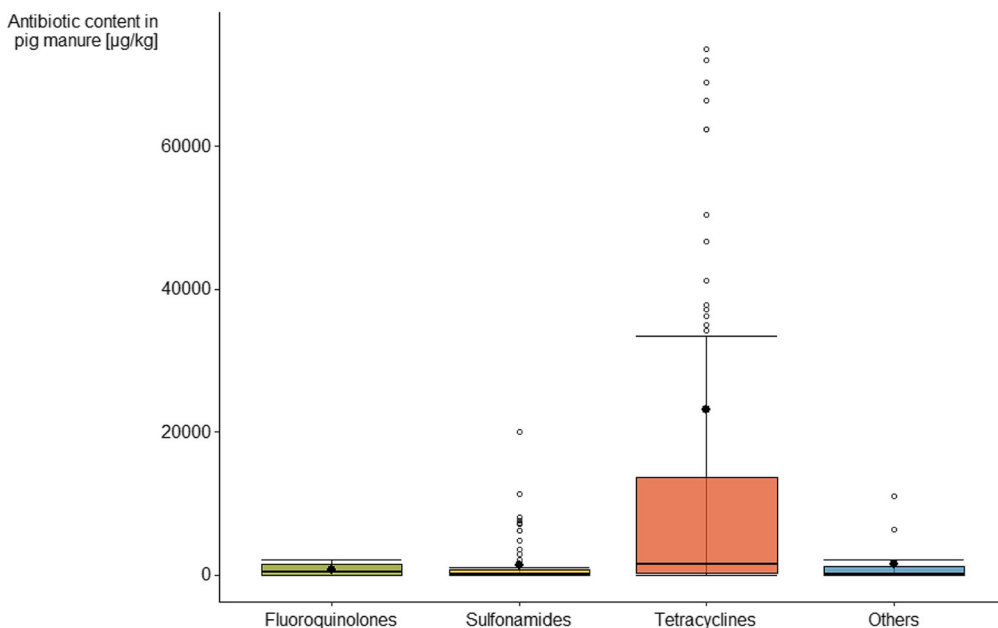


Fig. 1 Box and whisker plots showing the content of each antibiotics group in µg/kg (fluoroquinolones, sulfonamides, tetracyclines, and all other groups of antibiotics) detected in pig manure. Data were obtained from Berendsen et al. (2015), Chen et al. (2012), Cheng et al. (2016), Hamscher et al. (2005), Hanna et al. (2018), Hu et al. (2008), Jacobsen

and Halling-Sørensen (2006), Ji et al. (2012), Li et al. (2013), Martínez-Carballo et al. (2007), Qiao et al. (2012), Topi and Spahiu (2020), van den Meersche et al. (2016), Widyasari-Mehta et al. (2016), Xu et al. (2020), Zhang et al. (2015), and Zhao et al. (2010).

3 Results and discussion

3.1 Antibiotics content in pig manure and pig slurry

Overall, we found that pig manure contained antibiotics up to 376,210 µg/kg (tetracyclines group, Fig. 1). The highest median value for antibiotic content in pig manure was detected

for the tetracyclines group as well (median ± SE = 1,550 ± 6,209 µg/kg). The lowest median value for antibiotic content in pig manure was detected for the sulfonamides group (median ± SE = 98 ± 485 µg/kg, max 20,000 µg/kg). The average content for the fluoroquinolones and all other antibiotic groups were 465 ± 224 µg/kg, max 2,230 µg/kg, and 185 ± 1,075 µg/kg, max = 11,010 µg/kg, respectively.

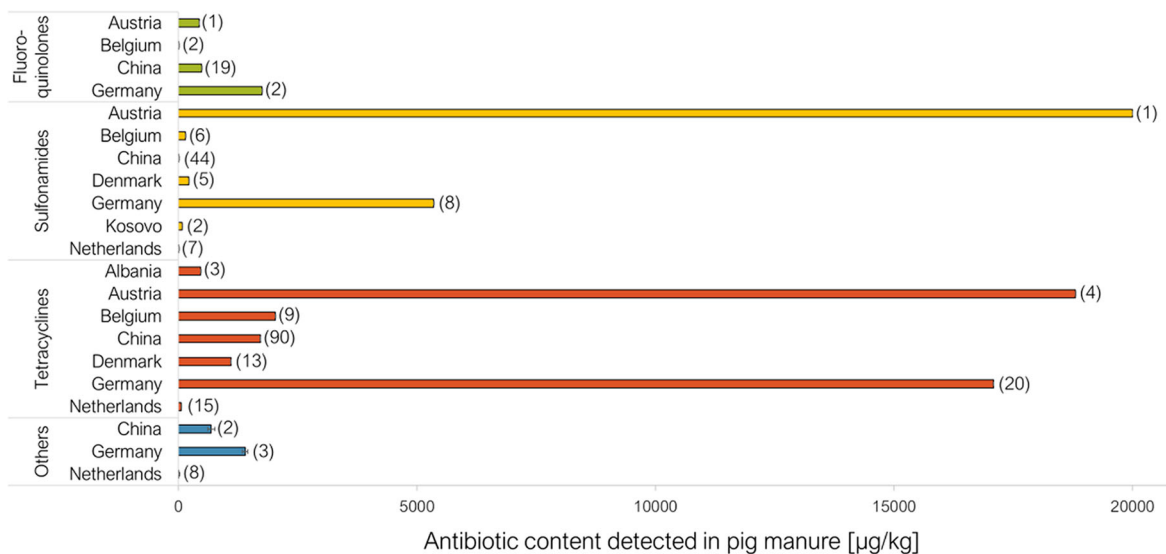
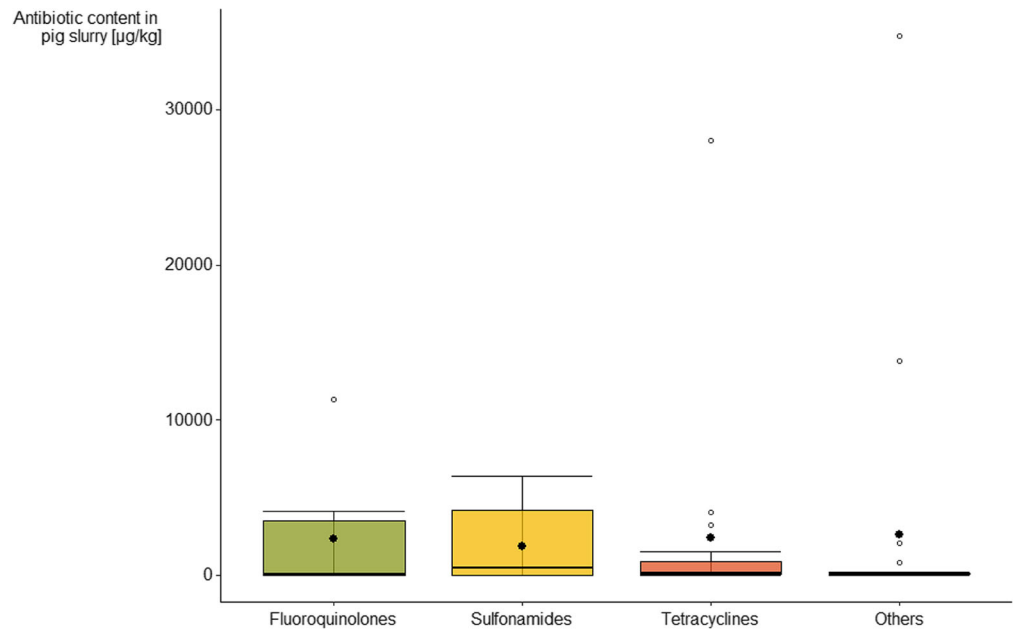


Fig. 2 Bar chart representing the median of antibiotics content and standard error (SE) of each antibiotics group detected in pig manure in different countries. The numbers in parentheses indicate the sample size of data used for the analysis (n). Data were obtained from Berendsen et al. (2015), Chen et al. (2012), Cheng et al. (2016), Hamscher et al. (2005),

Hanna et al. (2018), Hu et al. (2008), Jacobsen und Halling-Sørensen (2006), Ji et al. (2012), Li et al. (2013), Martínez-Carballo et al. (2007), Qiao et al. (2012), Topi und Spahiu (2020), van den Meersche et al. (2016), Widyasari-Mehta et al. (2016), Xu et al. (2020), Zhang et al. (2015), and Zhao et al. (2010).

Fig. 3 Box and whisker plots showing the content of each antibiotics group in $\mu\text{g}/\text{kg}$ (fluoroquinolones, sulfonamides, tetracyclines, and all other groups of antibiotics) detected in pig slurry. Data were obtained from Blackwell et al. (2009), Bourdat-Deschamps et al. (2014), Brambilla et al. (2007), Dreher et al. (2012), García-Sánchez et al. (2013), Haller et al. (2002), Hamscher et al. (2002), Rasschaert et al. (2020), Reichel et al. (2013), Sengeløv (2003), and Weiss et al. (2008).



A strong variation was found within the antibiotics content in pig manure among different countries (Fig. 2). Antibiotics of the fluoroquinolones group were detected in four countries (Austria, Belgium, China, and Germany). The median value of fluoroquinolones in pig manure ranged from 5 ± 0.04 (median \pm SE, Belgium, $n = 2$) to $1,755 \pm 1$ $\mu\text{g}/\text{kg}$ (median \pm SE, Germany, $n = 2$). Sulfonamides were detected in seven countries (Austria, Belgium, China, Denmark, Germany, Kosovo, and the Netherlands). Their content varied from 6 ± 0.2 (mean \pm SE, Netherlands, $n = 7$) to $20,000$ $\mu\text{g}/\text{kg}$ (Austria, $n = 1$). Antibiotics of the tetracyclines group were also detected in seven countries Albania, Austria, Belgium, China, Denmark, Germany, and the

Netherlands). The tetracyclines content varied from 61 ± 2 (median \pm SE, Netherlands, $n = 15$) to $18,803 \pm 2$ $\mu\text{g}/\text{kg}$ (median \pm SE, Austria, $n = 4$).

We found that pig slurry contained antibiotics up to $34,730$ $\mu\text{g}/\text{kg}$ (combination of other antibiotic groups, Fig. 3). The highest median value for antibiotic content in pig slurry was conducted for the sulfonamides group (median \pm SE = $490 \pm 1,018$ $\mu\text{g}/\text{kg}$, max = $6,400$ $\mu\text{g}/\text{kg}$). The lowest median value was found for fluoroquinolones in pig slurry (median \pm SE = $52 \pm 1,504$ $\mu\text{g}/\text{kg}$, max = $11,300$ $\mu\text{g}/\text{kg}$). The median content for the tetracyclines group and combination of other antibiotics were $95 \pm 1,018$ $\mu\text{g}/\text{kg}$, max $28,000$ $\mu\text{g}/\text{kg}$, and $58 \pm 2,287$ $\mu\text{g}/\text{kg}$, respectively.

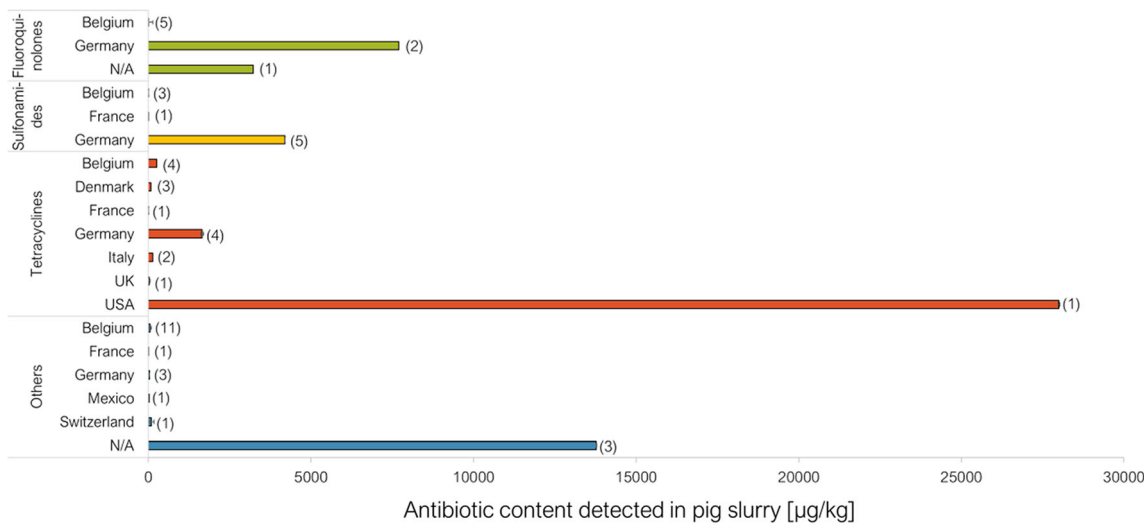


Fig. 4 Bar chart representing the median of antibiotics content and standard error (SE) of each antibiotics group detected in pig slurry in different countries. The numbers in parentheses indicate the sample size of data used for the analysis (n). Data were obtained from Blackwell et al.

(2009), Bourdat-Deschamps et al. (2014), Brambilla et al. (2007), Dreher et al. (2012), García-Sánchez et al. (2013), Haller et al. (2002), Hamscher et al. (2002), Rasschaert et al. (2020), Reichel et al. (2013), Sara et al. (2013), Sengeløv (2003), and Weiss et al. (2008).

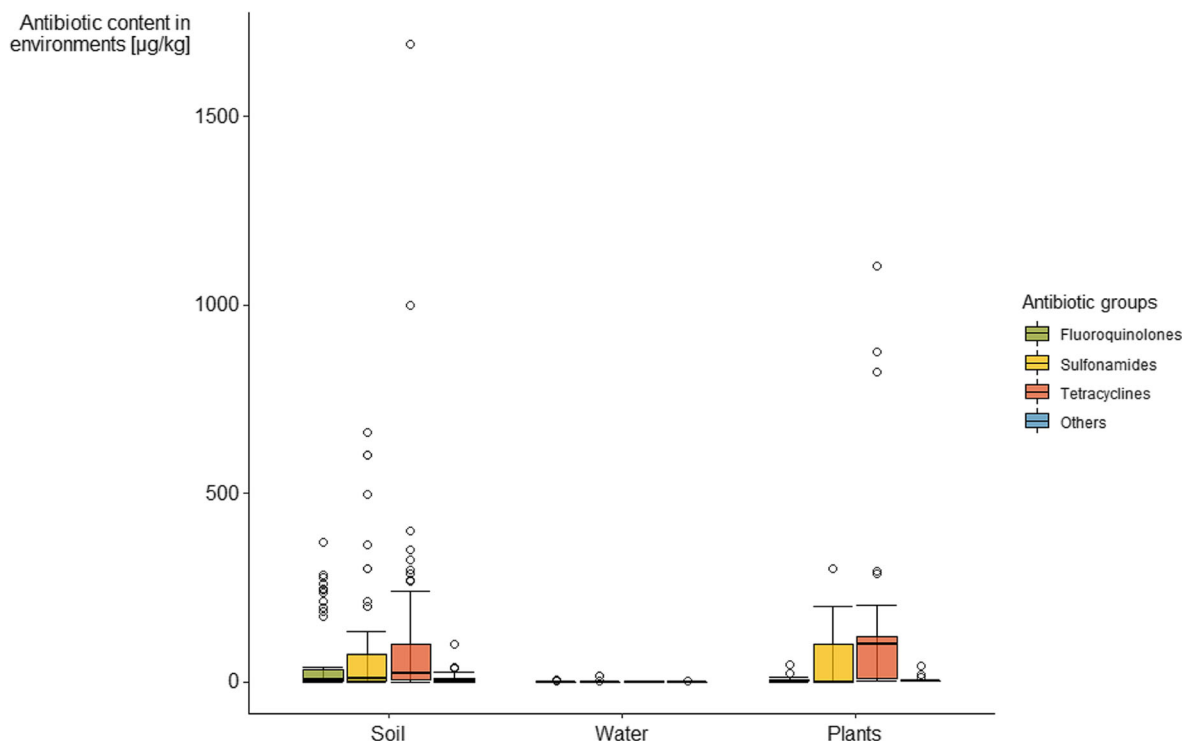


Fig. 5 Box and whisker plots showing the content of each antibiotics group (fluoroquinolones, sulfonamides, tetracyclines, and all other groups of antibiotics) in µg/kg for soil and plants, and in µg/L for water. Data were obtained from Blackwell et al. (2009), Camotti Bastos et al. (2018), Campagnolo et al. (2002), Chen et al. (2012), Cheng et al. (2016), Conde-Cid et al. (2018), Hamscher et al. (2005), Hamscher et al.

(2002), Hamscher and Mohring (2012), Hanna et al. (2018), Hu et al. (2010), Jacobsen et al. (2004), Kay et al. (2005), Martínez-Carballo et al. (2007), Marszałek et al. (2019), Qiao et al. (2012), Raich-Montiu et al. (2007), Rasschaert et al. (2020), Reichel et al. (2013), Sengelov (2003), Shelver et al. (2010), Weiss et al. (2008) and Zhang et al. (2015).

Within antibiotic groups detected in pig slurry, a strong variation was found (Fig. 4). The content of fluoroquinolones group ranged from 14.2 ± 13.8 (median \pm SE, Belgium, $n = 5$)

to $7,700 \pm 0.1$ µg/kg (median \pm SE, Germany, $n = 2$). Sulfonamides were detected in three countries in Europe (Belgium, France, and Germany). The contents of sulfonamides

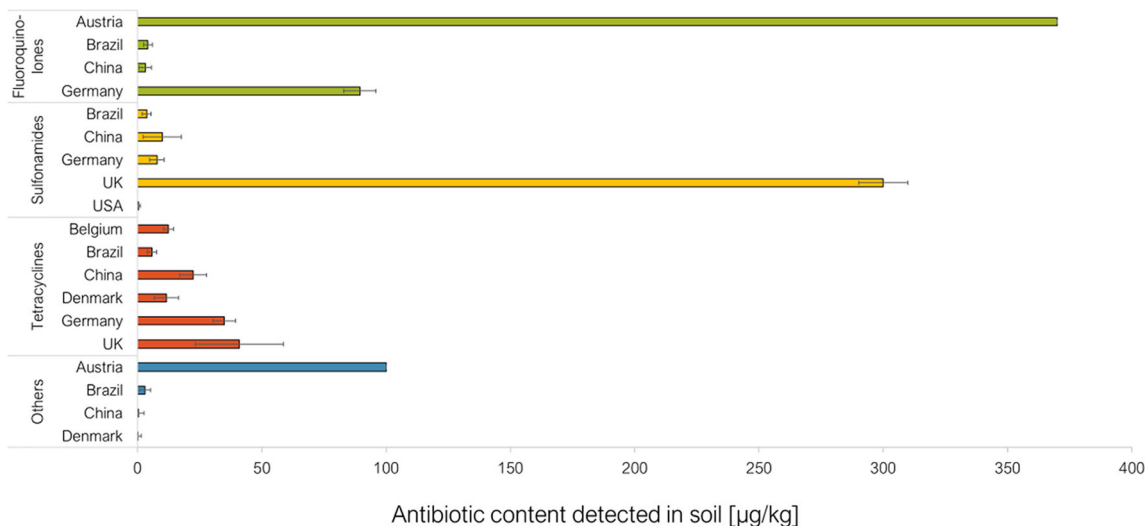


Fig. 6 Bar chart representing the median content and standard error (SE) of each antibiotics group detected in soil in different countries. The numbers in parentheses indicate the sample size of data used for the analysis (n). Data were obtained from Blackwell et al. (2009), Camotti Bastos et al. (2018), Chen et al. (2012), Cheng et al. (2016), Hamscher

et al. (2002), Hamscher et al. (2005), Hanna et al. (2018), Hu et al. (2010), Kay et al. (2005), Marszałek et al. (2019), Martínez-Carballo et al. (2007), Rasschaert et al. (2020), Reichel et al. (2013), Sengelov (2003), Shelver et al. (2010), and Zhang et al. (2015).

varied from 0.7 (France, $n = 1$) to $4,200 \pm 0.1 \mu\text{g/kg}$ (median \pm SE, Germany, $n = 5$). Tetracyclines were detected in seven countries (Belgium, Denmark, France, Germany, Italy, UK, and USA). The highest content of tetracyclines was found in the USA with $28,000 \mu\text{g/kg}$ ($n = 1$), while the lowest content was found in the UK ($30 \mu\text{g/kg}$, $n = 1$).

3.2 Antibiotics content in soil, water, and plants

In the environment, we observed the highest amount of antibiotics in soil for tetracyclines with up to $1,691 \mu\text{g/kg}$, in water for sulfonamides with up to $16 \mu\text{g/L}$, and in plants for tetracyclines with up to $1,104 \mu\text{g/kg}$ (Fig. 5). The highest median value for antibiotic content was found for tetracyclines in soil and plants (median \pm SE = $21.2 \pm 26.8 \mu\text{g/kg}$, and $100 \pm 59.4 \mu\text{g/kg}$, respectively), and for sulfonamides in soil as well (median \pm SE = $9.5 \pm 17.8 \mu\text{g/kg}$). The lowest median content was found for the fluoroquinolones group in water (median \pm SE = $0.004 \pm 0.5 \mu\text{g/L}$ respectively).

In soil, the highest content of fluoroquinolones was detected in Austria ($370 \mu\text{g/kg}$, $n = 1$, Fig. 6), followed by Germany (median \pm SE = $90 \pm 7 \mu\text{g/kg}$, $n = 20$). Sulfonamides were detected in high content in agricultural soils in the UK (median \pm SE = $300 \pm 10 \mu\text{g/kg}$, $n = 9$), followed by China (median \pm SE = $10 \pm 7.7 \mu\text{g/kg}$, $n = 21$). The highest content of tetracyclines was detected in the UK (median \pm SE = $41 \pm 18 \mu\text{g/kg}$, $n = 9$), followed by Germany (median \pm SE = $35 \pm 5 \mu\text{g/kg}$, $n = 43$) and China (median \pm SE = $22 \pm 5 \mu\text{g/kg}$, $n = 35$). Trimethoprim was detected in Austria (median = $100 \mu\text{g/kg}$, $n = 1$) and a combination of other antibiotics were found in Brazil (median \pm SE = 3.1 ± 2.2), whereas in other

countries, the contents of other antibiotic groups were low (median $< 1 \mu\text{g/kg}$).

In water, high fluoroquinolones content was detected in water from field streams in the USA (median \pm SE = $3 \pm 0.7 \mu\text{g/L}$, $n = 3$, Fig. 7) followed by China (median \pm SE = $0.002 \pm 0.3 \mu\text{g/L}$, $n = 12$), including wastewater (max = $0.7 \mu\text{g/L}$), drinking water (max = $0.01 \mu\text{g/L}$), and river water (max = $0.004 \mu\text{g/L}$). High contents of sulfonamides were detected in surface and groundwater in the USA (median \pm SE = $0.3 \pm 0.8 \mu\text{g/L}$, max = $3 \mu\text{g/L}$, $n = 7$) and Germany (median \pm SE = $0.23 \pm 1.98 \mu\text{g/L}$, max = $15 \mu\text{g/L}$, $n = 8$). Contents of tetracyclines in surface water were under $2 \mu\text{g/L}$, with the highest content detected in farm springs, field wells or tile lines in the USA (median \pm SE = $1.3 \pm 0.6 \mu\text{g/L}$, $n = 6$), followed by Germany (median \pm SE = $0.1 \pm 0.6 \mu\text{g/L}$, $n = 7$) and China (median \pm SE = $0.1 \pm 0.5 \mu\text{g/L}$, $n = 10$). The highest amount for other antibiotics was detected for lincomycin in surface water in Germany (max = $0.7 \mu\text{g/L}$), and groundwater in USA (max = $1.4 \mu\text{g/L}$).

In plants, fluoroquinolones were detected in China (median \pm SE = $1.4 \pm 2.6 \mu\text{g/kg}$, $n = 11$, Fig. 8), wherein the highest content was found in vegetables (max = $45 \mu\text{g/kg}$). The highest content of sulfonamides was found in grass and corn in Spain (median \pm SE = $100 \pm 6 \mu\text{g/kg}$, $n = 9$, max = $200 \mu\text{g/kg}$), whereas the leaves, roots, and stalk of radish, rape, celery, and coriander in China contained a maximum of $0.8 \mu\text{g/kg}$ (median \pm SE = $0.3 \pm 0.4 \mu\text{g/kg}$, $n = 15$). The highest tetracyclines contents were detected in wheat roots, stems and leaves, and grain in Germany (median \pm SE = $822 \pm 18 \mu\text{g/kg}$, $n = 5$, max = $1,104 \mu\text{g/kg}$), followed by grass and corn in Spain (median \pm SE = $100 \pm 4 \mu\text{g/kg}$, $n = 9$, max = $200 \mu\text{g/kg}$) and leaves, roots, and stalk of radish, rape, celery,

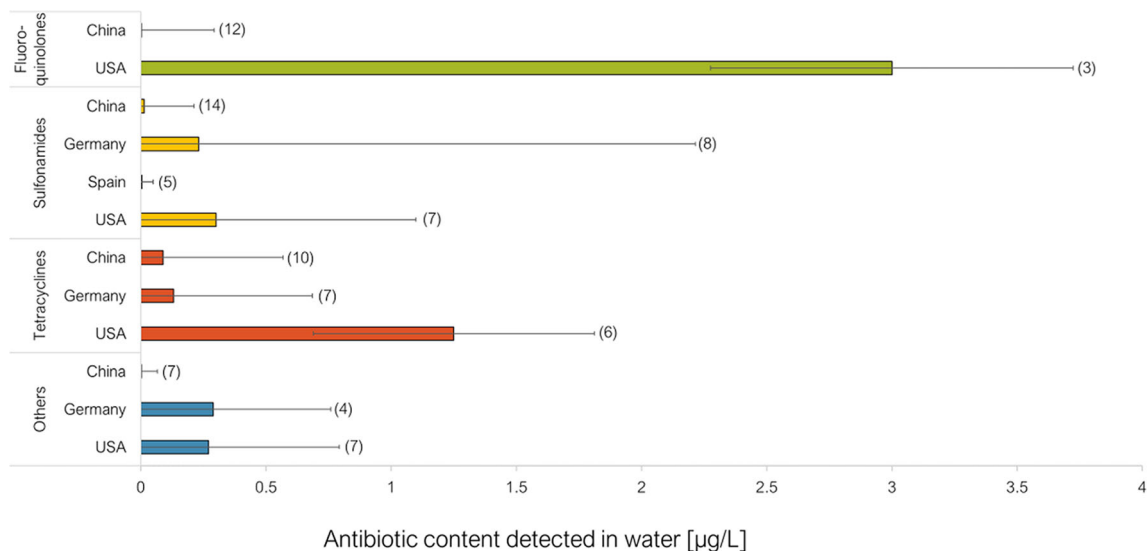


Fig. 7 Bar chart representing the median content and standard error (SE) of each antibiotics group detected in water in different countries. The numbers in parentheses indicate the sample size of data used for the analysis (n). Data were obtained from Campagnolo et al. (2002), Cheng

et al. (2016), Hamscher und Mohring (2012), Hanna et al. (2018), Hu et al. (2010), Marszałek et al. (2019), Raich-Montiu et al. (2007), Shelver et al. (2010), and Weiss et al. (2008).

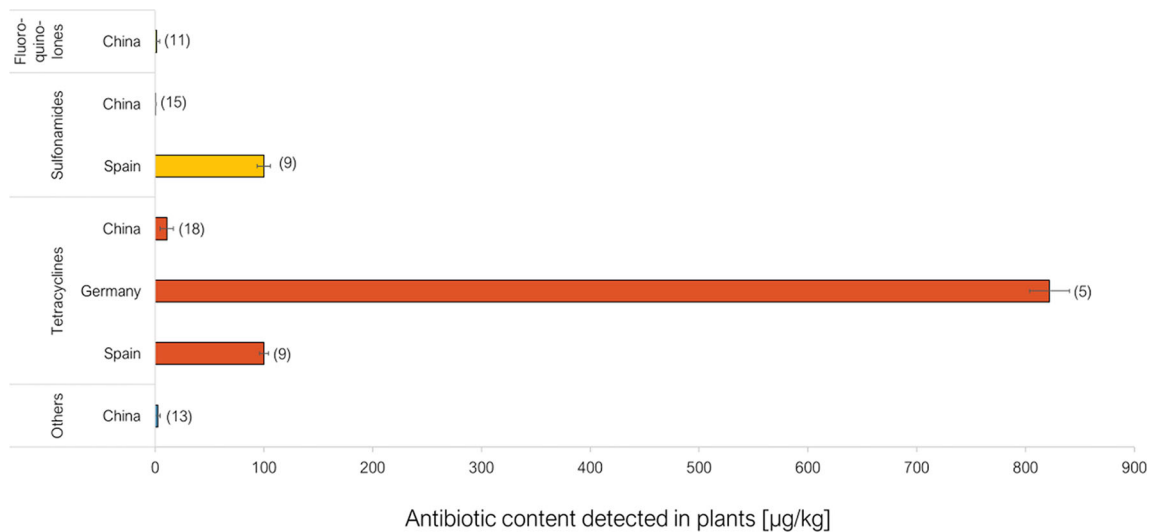


Fig. 8 Bar chart representing the median content and standard error (SE) of each antibiotics group detected in plants in different countries. The numbers in parentheses indicates the sample size of data used for the

analysis (n). Data were obtained from Conde-Cid et al. (2018), Grote et al. (2007), Hanna et al. (2018), and Hu et al. (2010).

and coriander in China (median \pm SE = 10.7 ± 6.1 $\mu\text{g}/\text{kg}$, $n = 9$, max = 200 $\mu\text{g}/\text{kg}$). The highest content among other antibiotics was detected for chloramphenicol in vegetables in China (max = 41 $\mu\text{g}/\text{kg}$).

3.3 Discussion

Veterinary antibiotics in pig manure and slurry were detected in 11 European countries (Albania, Austria, Belgium, Denmark, France, Germany, Italy, Kosovo, Netherlands, Switzerland, and the UK), two countries in America (Mexico and the USA), and one in Asia (China). Among different veterinary antibiotics, especially fluoroquinolones, sulfonamides, and tetracyclines were detected in pig manure and pig slurry. The content of each group of veterinary antibiotics varied greatly in different locations in which they were detected. Pig manure from Austria and Germany (sulfonamides, tetracyclines), Belgium and China (tetracyclines) as well as slurry from the USA (tetracyclines) and Germany (fluoroquinolones, sulfonamides, and tetracyclines) contained high amounts of different antibiotics groups. The variation of detected antibiotics is due to the allowed application amount of veterinary antibiotics in different countries (Marszałek et al. 2019). Furthermore, different antibiotics may have different degradation mechanisms and thus different half-life times (Blackwell et al. 2004). The selected studies also have different sampling times (most are not given in the study). Thus, some antibiotics, which further degraded over time, can potentially be detected in low amounts.

Moreover, antibiotics used in animal husbandry are found to contaminate surrounding environments. This includes soil and water bodies as well as the accumulation in plants. Although antibiotics become much less stable after being

applied onto agricultural land (Marszałek et al. 2019; Venglovsky et al. 2009), soil fertilized with contaminated animal excreta in form of manure or slurry was found to contain up to ten times (tetracyclines in the UK, max = 1000 $\mu\text{g}/\text{L}$, Kay et al. (2005)) the proposed EU threshold value of the PEC of veterinary antibiotics in soil ($\text{PEC}_{\text{soil}} = 100$ $\mu\text{g}/\text{kg}$, Committee for Medicinal Products for Veterinary Use (2016)). Soils in Austria, China, Germany, and the UK contained highest amounts of different antibiotic groups among other countries. Due to surface runoff or infiltration, water bodies showed concentrations of antibiotic substances with more than 30 times the amount of the PEC for surface water (tetracyclines in the UK, max = 36 $\mu\text{g}/\text{L}$, Blackwell et al. (2005)) and three times the amount of the PEC for groundwater (sulfonamides in Germany, max = 0.4 $\mu\text{g}/\text{L}$, Hamscher and Mohring (2012)) according to the EU directive ($\text{PEC}_{\text{groundwater}} = 0.1$ $\mu\text{g}/\text{L}$, $\text{PEC}_{\text{surfacewater}} = 1$ $\mu\text{g}/\text{L}$, Committee for Medicinal Products for Veterinary Use (2016)). However, antibiotics can degrade very quickly when they are distributed into the environment or have a high affinity for adsorption onto soil organic matter (Kemper et al. 2008). Hereby tetracyclines, which are the most commonly used veterinary antibiotic in the EU, bind strongly onto soil organic matter due to complexes with double-charged ions. Consequently, low concentrations of this antibiotic group can be found in water which coincides with the results of our analysis (Kay et al. 2004; Kemper et al. 2008). For other groups, it can be assumed that the concentration in the environment varies depending on the amount of time manure and slurry were stored before their contribution on agricultural lands (Kemper et al. 2008). Additionally, these substances can eventually reach the food chain due to root uptake and thus be detected in crops and vegetables growing directly

adjacent to the fertilized soil (Hamscher and Mohring 2012; Marszałek et al. 2019). This report detected antibiotics concentrations in plants ranging from 0.1 µg/kg (Hu et al. 2008) up to 1,104 µg/kg (Grote et al. 2007). And the maximum amount of antibiotics residues allowed in animal products is 100–600 µg/kg (The European Commission 2010). Therefore, the detected content of antibiotics in plants exceeds the EU regulations for animal food products by up to ten-fold. Moreover, as slurry contains more water than manure, its mobility after application leads to the spreading of contained pathogens, such as bacteria or viruses, into soil. Through transportation of these pathogens deeper into soil and water bodies they can survive over a long period of time (up to 119 days in soil (*Brachyspira pilosicoli*), Marszałek et al. (2019)). This creates the risk of animal pathogens, including antibiotic-resistant bacteria, reaching the groundwater (Marszałek et al. 2019; Venglovsky et al. 2009).

In this study, literature reporting antibiotics in manure and slurry were selected. Different types of antibiotics were found mainly in the European countries. To identify the specificity of antibiotics residues to country or continent, the information of antibiotics detected in slurry and manure from many more countries should be included. Antibiotics residues in the environments were detected in Asia (China), Europe (Austria, Belgium, Denmark, Germany, Spain, and the UK) and America (Brazil and the USA). In soils, fluoroquinolones, sulfonamides, and tetracyclines were measured in certain amounts in many countries across all three continents and thus no specificity of antibiotic residues to country or continent can be observed. Nevertheless, sulfonamides and tetracyclines in Germany were detected in high content in plants and water, and fluoroquinolones in the USA in water whereas their concentrations are much lower in other countries.

4 Conclusion

We conclude that residues of antibiotics used in agricultural livestock can be found in manure and slurry which is used as a natural fertilizer. We also confirm that these antibiotics residues contaminated the environment (including soil, water, and plants) directly adjacent to agricultural land fertilized with slurry. However, since only little data is available from non-European countries, we were not able to test or observe a significant specificity of these antibiotic residues to country or continent.

The large amounts of organic matter and nutrients in livestock manure and slurry has brought many positive effects as a natural fertilizer, such as improving soil as well as enhancing crop yield and quality. However, the intensive and excessive use of veterinary antibiotics in livestock agriculture has led to several environmental problems due to the absence of application limits and hygienization processes (Hamscher and

Mohring 2012; Marszałek et al. 2019). To avoid these substances as well as the resulting drug-resistant bacteria to reach groundwater or crops, it is necessary to minimize environmental contamination. The overall use of veterinary antibiotics needs to be reduced in order to diminish the amount of drugs in manure and slurry which is eventually used as fertilizer. There are different treatment techniques which can reduce the concentration of antibiotics in animal excrements (see Marszałek et al. (2019) and the references therein): anaerobic digestion, aeration, electron beam irradiation, and sorption. Consequently, the proven partial or almost complete elimination of antibiotics in slurry shows the need for a wider contribution of these techniques on a commercial scale.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s13593-022-00762-y>.

Code availability Available upon request to the corresponding author

Authors' contributions LF and BT conducted the study including literature search, data evaluation, and manuscript writing, BG improved the manuscript and had the idea for the study.

Funding Open Access funding enabled and organized by Projekt DEAL.

Data availability Available upon request to the corresponding author

Declarations

Ethics approval Not applicable

Consent to participate Not applicable

Consent for publication Not applicable

Conflict of interests The authors declare no competing interests.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- Berendsen BJA, Wegh RS, Memelink J, Zuidema T, Stolker LAM (2015) The analysis of animal faeces as a tool to monitor antibiotic usage. *Talanta* 132:258–268. <https://doi.org/10.1016/j.talanta.2014.09.022>
- Blackwell PA, Boxall ABA, Kay P, Noble H (2005) Evaluation of a lower tier exposure assessment model for veterinary

- medicines. *J Agr Food Chem* 53:2192–2201. <https://doi.org/10.1021/jf049527b>
- Blackwell PA, Holten LHC, Ma HP, Halling-Sørensen B, Boxall ABA, Kay P (2004) Ultrasonic extraction of veterinary antibiotics from soils and pig slurry with SPE clean-up and LC-UV and fluorescence detection. *Talanta* 64:1058–1064. <https://doi.org/10.1016/j.talanta.2004.05.006>
- Blackwell PA, Kay P, Ashauer R, Boxall BA (2009) Effects of agricultural conditions on the leaching behaviour of veterinary antibiotics in soil. *Chemosphere* 75(1):13–19. <https://doi.org/10.1016/j.chemosphere.2008.11.070>
- Bohrer REG, Carissimi E, Lopez DAR, Wolff DB, Da Silva DM, Prestes OD (2019) Compostagem de efluente suíno no tratamento de resíduos de fármacos veterinários. *SCA* 40:2813. <https://doi.org/10.5433/1679-0359.2019v40n6p2813>
- Bourdat-Deschamps M, Leang S, Bernet N, Daudin JJ, Nélieu S (2014) Multi-residue analysis of pharmaceuticals in aqueous environmental samples by online solid-phase extraction-ultra-high-performance liquid chromatography-tandem mass spectrometry: optimisation and matrix effects reduction by quick, easy, cheap, effective, rugged and safe extraction. *J Chromatogr A* 1349:11–23. <https://doi.org/10.1016/j.chroma.2014.05.006>
- Brambilla G, Patrizzii M, de Filippis SP, Bonazzi G, Mantovi P, Barchi D, Migliore L (2007) Oxytetracycline as environmental contaminant in arable lands. *Analytica chimica acta* 586(1-2):326–329. <https://doi.org/10.1016/j.aca.2006.11.019>
- Camotti Bastos M, Rheinheimer dos Santos D, Aubertheau E, Le Guet T, Caner L et al (2018) Antibiotics and microbial resistance in Brazilian soils under manure application. *Land Degrad Dev* 29(8):2472–2484. <https://doi.org/10.1002/ldr.2964>
- Campagnolo E, Johnson KR, Karpati A, Rubin CS, Kolpin DW, Meyer MA et al (2002) Antimicrobial residues in animal waste and water resources proximal to large-scale swine and poultry feeding operations. *Sci Total Environ* 299(1-3):89–95. [https://doi.org/10.1016/S0048-9697\(02\)00233-4](https://doi.org/10.1016/S0048-9697(02)00233-4)
- Chee-Sanford JC, Mackie RL, Koike S, Krapac IG, Lin YF, Yannarell AC, Maxwell S, Aminov RI (2009) Fate and transport of antibiotic residues and antibiotic resistance genes following land application of manure waste. *J Environ Qual* 38:1086–1108. <https://doi.org/10.2134/jeq2008.0128>
- Chen YS, Zhang HB, Luo YM, Song J (2012) Occurrence and assessment of veterinary antibiotics in swine manures: A case study in East China. *Chin Sci Bull* 57:606–614. <https://doi.org/10.1007/s11434-011-4830-3>
- Cheng W, Li J, Wu Y, Xu L, Su C, Qian Y, Zhu YG, Chen H (2016) Behavior of antibiotics and antibiotic resistance genes in eco-agricultural system: A case study. *J Hazard Mater* 304:18–25. <https://doi.org/10.1016/j.jhazmat.2015.10.037>
- Committee for Medicinal Products for Veterinary Use (2016) Guideline on environmental impact assessment for VMPs in support of the VICH GL6 and GL38. EMA/CVMP/ERA/418282/2005-Rev.1-Corr. European Medicines Agency.
- Conde-Cid M, Álvarez-Esmoris C, Paradelo-Núñez R, Nóvoa-Muñoz JC, Arias-Estévez M, Álvarez-Rodríguez E, Fernández-Sanjurjo MJ, Núñez-Delgado A (2018) Occurrence of tetracyclines and sulfonamides in manures, agricultural soils and crops from different areas in Galicia (NW Spain). *J Clean Prod* 197:491–500. <https://doi.org/10.1016/j.jclepro.2018.06.217>
- Destoumieux-Garzón D, Mavingui P, Boetsch G, Boissier J, Darriet F, Duboz P, Fritsch C, Giraudoux P, le Roux F, Morand S, Paillard C, Pontier D, Sueur C, Voituron Y (2018) The One Health Concept: 10 Years Old and a Long Road Ahead. *Front Vet Sci* 5:14. <https://doi.org/10.3389/fvets.2018.00014>
- Dreher TM, Mott HV, Lupo CD, Oswald AS, Clay SA, Stone JJ (2012) Effects of chlortetracycline amended feed on anaerobic sequencing batch reactor performance of swine manure digestion. *Bioresour Technol* 125:65–74. <https://doi.org/10.1016/j.biortech.2012.08.077>
- Fox J, Weisberg S (2019) *An R Companion to Applied Regression*. Edited by Sage Available online at <https://socialsciences.mcmaster.ca/jfox/Books/Companion/>. Accessed 12 Dec 2021
- García-Sánchez L, Garzón-Zúñiga MA, Buelna G, Moeller-Chávez GE, Noyola A, Avilez-Flores M, Estrada-Arriaga EB (2013) Occurrence of tylosin in swine wastewater in Mexico. *Water Sci Technol* 68(4):894–900. <https://doi.org/10.2166/wst.2013.323>
- Grote M, Schwake-Anduschus C, Michel R, Stevens H, Heyser W, Langenkämper G et al (2007) Incorporation of veterinary antibiotics into crops from manured soil. *Landbauforschung Völkenrode* 57:25–32
- Haller MY, Müller SR, McArdell CS, Alder AC, Suter MJF (2002) Quantification of veterinary antibiotics (sulfonamides and trimethoprim) in animal manure by liquid chromatography– mass spectrometry. *J Chromatogr A* 952:111–120
- Hamscher G, Mohring SAI (2012) Tierarzneimittel in Böden und in der aquatischen Umwelt. *Chemie Ingenieur Technik* 84:1052–1061. <https://doi.org/10.1002/cite.201100255>
- Hamscher G, Pawelzick HT, Höper H, Nau H (2005) Different behavior of tetracyclines and sulfonamides in sandy soils after repeated fertilization with liquid manure. *Environ Toxicol Chem* 24:861–868. <https://doi.org/10.1897/04-182r.1>
- Hamscher G, Sczesny S, Höper H, Nau H (2002) Determination of persistent tetracycline residues in soil fertilized with liquid manure by high-performance liquid chromatography with electrospray ionization tandem mass spectrometry. *Analytical chemistry* 74(7):1509–1518. <https://doi.org/10.1021/ac015588m>
- Hanna N, Sun P, Sun Q, Li X, Yang X, Ji X, Zou H, Ottoson J, Nilsson LE, Berglund B, Dyar OJ, Tamhankar AJ, Stålsby Lundborg C (2018) Presence of antibiotic residues in various environmental compartments of Shandong province in eastern China: Its potential for resistance development and ecological and human risk. *Environ Int* 114:131–142. <https://doi.org/10.1016/j.envint.2018.02.003>
- Hu X, Xian-Gang LY, Zhou Q, Qi-Xing X (2008) Determination of Thirteen Antibiotics Residues in Manure by Solid Phase Extraction and High Performance Liquid Chromatography. *Chinese J Anal Chem* 34:1162–1166. [https://doi.org/10.1016/S1872-2040\(08\)60063-8](https://doi.org/10.1016/S1872-2040(08)60063-8)
- Hu X, Zhou Q, Luo Y (2010) Occurrence and source analysis of typical veterinary antibiotics in manure, soil, vegetables and groundwater from organic vegetable bases, northern China. *Environ Pollut* 158(9):2992–2998. <https://doi.org/10.1016/j.envpol.2010.05.023>
- Jacobsen AM, Halling-Sørensen B (2006) Multi-component analysis of tetracyclines, sulfonamides and tylosin in swine manure by liquid chromatography-tandem mass spectrometry. *Anal Bioanal Chem* 384:1164–1174. <https://doi.org/10.1007/s00216-005-0261-9>
- Jacobsen AM, Halling-Sørensen B, Ingerslev F, Honoré Hansen S (2004) Simultaneous Extraction of Tetracycline, Macrolide and Sulfonamide Antibiotics from Agricultural Soils Using Pressurised Liquid Extraction, Followed by Solid-Phase Extraction and Liquid Chromatography-Tandem Mass Spectrometry. *J Chromatogr A* 1038:157–170
- Ji X, Shen Q, Liu F, Ma J, Xu G, Wang Y, Wu M (2012) Antibiotic resistance gene abundances associated with antibiotics and heavy metals in animal manures and agricultural soils adjacent to feedlots in Shanghai, China. *J Hazard Mater* 235-236:178–185. <https://doi.org/10.1016/j.jhazmat.2012.07.040>
- Kay P, Blackwell PA, Boxall ABA (2005) Column studies to investigate the fate of veterinary antibiotics in clay soils following slurry application to agricultural land. *Chemosphere* 60:497–507. <https://doi.org/10.1016/j.chemosphere.2005.01.028>
- Kay P, Blackwell PA, Boxall ABA (2004) Fate of veterinary antibiotics in a macroporous tile drained clay soil. *Environ Toxicol Chem* 23:1136–1144. <https://doi.org/10.1897/03-374>

- Kemper N, Färber H, Skutlarek D, Krieter J (2008) Analysis of antibiotic residues in liquid manure and leachate of dairy farms in Northern Germany. *Agr Water Manage* 95:1288–1292. <https://doi.org/10.1016/j.agwat.2008.05.008>
- Kivits T, Broers HP, Beeltje H, van Vliet M, Griffioen J (2018) Presence and fate of veterinary antibiotics in age-dated groundwater in areas with intensive livestock farming. *Environ Pollut* 241:988–998. <https://doi.org/10.1016/j.envpol.2018.05.085>
- Li YX, Zhang XL, Li W, Lu XF, Liu B, Wang J (2013) The residues and environmental risks of multiple veterinary antibiotics in animal faeces. *Environ Monit Assess* 185:2211–2220. <https://doi.org/10.1007/s10661-012-2702-1>
- Marszałek M, Kowalski Z, Makara A (2019) The Possibility of Contamination of Water-Soil Environment as a Result of the Use of Pig Slurry. *Ecol Chem Eng S* 26:313–330. <https://doi.org/10.1515/eces-2019-0022>
- Martínez-Carballo E, González-Barreiro C, Scharf S, Gans O (2007) Environmental monitoring study of selected veterinary antibiotics in animal manure and soils in Austria. *Environ Pollut* 148:570–579. <https://doi.org/10.1016/j.envpol.2006.11.035>
- Qiao M, Chen W, Su J, Zhang B, Zhang C (2012) Fate of tetracyclines in swine manure of three selected swine farms in China. *J Environ Sci* 24:1047–1052. [https://doi.org/10.1016/S1001-0742\(11\)60890-5](https://doi.org/10.1016/S1001-0742(11)60890-5)
- Raich-Montiu J, Folch J, Compañó R, Granados M, Prat MD (2007) Analysis of trace levels of sulfonamides in surface water and soil samples by liquid chromatography-fluorescence. *J Chromatogr A* 1172(2):186–193. <https://doi.org/10.1016/j.chroma.2007.10.010>
- Rasschaert G, van Elst D, Colson L, Herman L, Ferreira HCC, Dewulf J et al (2020) Antibiotic Residues and Antibiotic-Resistant Bacteria in Pig Slurry used to Fertilize Agricultural Fields. *Antibiotics* (Basel, Switzerland) 9(1):34. <https://doi.org/10.3390/antibiotics9010034>
- Reichel R, Rosendahl I, Peeters ETHM, Focks A, Groeneweg J, Bierl R, Schlichting A, Amelung W, Thiele-Bruhn S (2013) Effects of slurry from sulfadiazine- (SDZ) and difloxacin- (DIF) medicated pigs on the structural diversity of microorganisms in bulk and rhizosphere soil. *Soil Biol Biochem* 62:82–91. <https://doi.org/10.1016/j.soilbio.2013.03.007>
- Sengeløv G (2003) Bacterial antibiotic resistance levels in Danish farmland as a result of treatment with pig manure slurry. *Environ Int* 28(7):587–595. [https://doi.org/10.1016/S0160-4120\(02\)00084-3](https://doi.org/10.1016/S0160-4120(02)00084-3)
- Shelver WL, Hakk H, Larsen GL, DeSutter TM, Casey FXM (2010) Development of an Ultra-High-Pressure Liquid Chromatography-Tandem Mass Spectrometry Multi-Residue Sulfonamide Method and Its Application to Water, Manure Slurry, and Soils from Swine Rearing Facilities. *J Chromatogr A* 1217:1273–1282
- The European Commission (2010) Commission Regulation (EU) No 37/2010 of 22 December 2009 on pharmacologically active substances and their classification regarding maximum residue limits in food-stuffs of animal origin. Official Journal of the European Union.
- Topi D, Spahiu J (2020) Presence of veterinary antibiotics in livestock manure in two Southeastern Europe countries, Albania and Kosovo. *Environ Sci Pollut R* 27:44552–44560. <https://doi.org/10.1007/s11356-020-10341-x>
- van den Meersche T, van Pamel E, van Poucke C, Herman L, Heyndrickx M, Rasschaert G, Daeseleire E (2016) Development, validation and application of an ultra high performance liquid chromatographic-tandem mass spectrometric method for the simultaneous detection and quantification of five different classes of veterinary antibiotics in swine manure. *J Chromatogr A* 1429:248–257. <https://doi.org/10.1016/j.chroma.2015.12.046>
- Venglovsky J, Sasakova N, Placha I (2009) Pathogens and antibiotic residues in animal manures and hygienic and ecological risks related to subsequent land application. *Bioresource Technol* 100:5386–5391. <https://doi.org/10.1016/j.biortech.2009.03.068>
- Weiss K, Schüssler W, Porzelt M (2008) Sulfamethazine and flubendazole in seepage water after the sprinkling of manured areas. *Chemosphere* 72(9):1292–1297. <https://doi.org/10.1016/j.chemosphere.2008.04.053>
- Wickham H (2009) ggplot2: Elegant Graphics for Data Analysis. Edited by Springer-Verlag New York Available online at <http://ggplot2.org>. Accessed 12 Dec 2021
- Widyasari-Mehta A, Hartung S, Kreuzig R (2016) From the application of antibiotics to antibiotic residues in liquid manures and digestates: A screening study in one European center of conventional pig husbandry. *J Environ Manage* 177:129–137. <https://doi.org/10.1016/j.jenvman.2016.04.012>
- Xia X, Wang Z, Fu Y, Du XD, Gao B, Zhou Y et al (2019) Association of colistin residues and manure treatment with the abundance of mcr-1 gene in swine feedlots. *Environ Int* 127:361–370. <https://doi.org/10.1016/j.envint.2019.03.061>
- Xu M, Li H, Li S, Li C, Li J, Ma Y (2020) The presence of tetracyclines and sulfonamides in swine feeds and feces: dependence on the antibiotic type and swine growth stages. *Environ Sci Pollut R* 27:43093–43102. <https://doi.org/10.1007/s11356-020-10266-5>
- Zhang HB, Luo YM, Wu L, Huang Y, Christie P (2015) Residues and potential ecological risks of veterinary antibiotics in manures and composts associated with protected vegetable farming. *Environ Sci Pollut R* 22:5908–5918. <https://doi.org/10.1007/s11356-014-3731-9>
- Zhao L, Dong YH, Wang H (2010) Residues of veterinary antibiotics in manures from feedlot livestock in eight provinces of China. *Sci Total Environ* 408:1069–1075. <https://doi.org/10.1016/j.scitotenv.2009.11.014>

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.