Persistence of Antibiotics in the Natural Environment: Scoping Review Protocol

Irene Bueno[1*], Huan He[2], André J. Nault[3], Sarah Ziemann[4], Lauren Degn[4], William A. Arnold[2a], Kristine H. Wammer[4a]

1. Department of Veterinary and Biomedical Sciences, University of Minnesota. 1971 Commonwealth Ave. Saint Paul, MN 55108. United States.
2. Department of Civil, Environmental, and Geo- Engineering, University of Minnesota. 500 Pillsbury Drive S.E. Minneapolis, MN 55455-0116. United States.
3. Health Sciences Libraries, University of Minnesota. 1988 Fitch Avenue. Saint Paul, MN 55108. United States.
4. Department of Chemistry, College of Arts & Sciences, University of St. Thomas. 2115 Summit Ave. Saint Paul, MN 55105. United States.

a: Co-senior authors

*Corresponding author: Irene Bueno (bueno004@umn.edu)

Abstract

Background: Antibiotics and their metabolites released into aquatic and soil environments have the potential to affect their microbial communities and can be a selection pressure to drive antimicrobial resistance emergence and spread. However, data about the persistence of antibiotics and metabolites into the natural environment are still lacking.

Objectives: The goal of this manuscript is to describe the protocol that will be used to conduct a formal scoping review of the current literature to address the following question: "*What is known*

from the existing literature about degradation of a selected group of antibiotic compounds in water, sediment, and soil?".

Eligibility criteria: Eligible studies will be primary research, in English, from any geographic location, published between 2000-2020, include water, sediment, and/or soil samples, were conducted in natural systems and/or laboratory studies with relevant data applied to natural systems, include data related to transformation by sunlight, biodegradation, and/or sorption processes, and include data for any of the following compounds: i) sulfonamides: sulfachlorpyridazine, sulfadiazine, sulfadimethoxine, sulfamethazine, sulfamethoxazole, sulfapyridine; ii) macrolides: erythromycin, roxithromycin, tylosin, azithromycin; iii) tetracyclines: chlortetracycline, doxycycline, oxytetracycline, tetracycline; iv) fluoroquinlones: ciprofloxacin, enrofloxacin, norfloxacin, ofloxacin; v) beta-lactams: penicillins; vi) others: carbadox, trimethoprim, lincomycin.

Sources of evidence: The following databases will be searched: Pubmed/MEDLINE, CAB Abstracts, Web of Science, and Scopus from 2000-2020.

Charting methods: We will extract data on general and specific study characteristics, study systems, and transformation processes/antibiotic compounds combinations.

Introduction

Rationale

Antimicrobial resistance (AMR) is a well-recognized global public health threat (WHO, 2020). According to the Centers for Disease Control and Prevention (CDC), close to three million people in the U.S. develop antibiotic-resistant infections each year, and this leads to more than 35,000 deaths (CDC, 2019). As AMR increases, attention is being directed towards the role of the natural environment. Watersheds, in particular, are recipients of antibiotic residues, antimicrobial resistant bacteria (ARB), and antimicrobial resistance genes (ARG) released from effluents of anthropogenic sources (Bueno et al., 2017; Bueno et al., 2018; Huijbers et al., 2015). These sources include but are not limited to wastewater treatment plants, animal agriculture and aquaculture, ethanol plants, crop production, and pharmaceutical manufacturing plants. Antibiotics and their metabolites released from these sources might pose an indirect risk to human, animal, and ecosystem health by driving the emergence of antimicrobial resistance in environmental bacteria (Rahube et al., 2016).

Studying the fate of antibiotics and their metabolites in the natural environment through field work is challenging and limited by resources. To overcome this restraint, mathematical and geospatial modeling play an important role in predicting the fate of these compounds within the complexity and uncertainties of natural systems (Singer, et al., 2006; Spicknall, et al., 2013). To fully understand the persistence of antibiotics in the natural environment and fully parameterize mathematical and geospatial models, information on degradation rates for different compounds is needed. Relevant removal processes include transformation by sunlight, degradation by microorganisms, and sorption to settling particles (Kümmerer, 2009). For targeted antibiotic

compounds, a scoping review will be conducted to compile relevant biodegradation, sorption, and direct and indirect photolysis rate constants of the target antibiotics to assess which gaps are most critical to fill to inform future transformation studies. In addition, data from the scoping review will be used to parameterize antibiotic dissemination models in the natural environment.

Objectives

The objective of this protocol is to describe the methods that will be used to address the following review question: *What is known from the existing literature about degradation for a selected group of antibiotic compounds in water, sediment, and soil?* In particular, what is known about:

- *transformation by sunlight (direct and indirect photolysis rate constants)?*
- *degradation by microorganisms (biodegradation rate constants)?*
- *sorption to settling particles (partition coefficients as a function of particle organic carbon content, mineral composition, and pH)?*

Methods

Protocol and registration

This protocol was prepared using the Preferred Reporting Items for Systematic Reviews and Meta-Analyses Extension for Scoping Reviews (PRISMA-ScR) (Tricco et al., 2018) and it will be published in the University of Minnesota Libraries Digital Conservancy (https://conservancy.umn.edu/).

Eligibility criteria

Studies are eligible for this review if they meet the following criteria:

a) Primary research.

b) Published in English.

c) Published between 2000-2020. The year 2000 was chosen because it was when analytical methods became sensitive enough to quantify antibiotic's environmentally-relevant concentrations.

d) Conducted in natural systems and/or in laboratory settings that provide relevant data to the natural systems.

e) Investigate any of the following transformation studies: sorption, biodegradation, and/or photolysis.

f) Investigate any of the following compounds: i) sulfonamides: sulfachlorpyridazine, sulfadiazine, sulfadimethoxine, sulfamethazine, sulfamethoxazole, sulfapyridine; ii) macrolides: erythromycin, roxithromycin, tylosin, azithromycin; iii) tetracyclines: chlortetracycline, doxycycline, oxytetracycline, tetracycline; iv) fluoroquinlones: ciprofloxacin, enrofloxacin, norfloxacin, ofloxacin; v) beta-lactams: penicillins (broadly); vi) others: carbadox, trimethoprim, and/or lincomycin.

Information sources

The following databases will be searched for relevant studies: Pubmed/MEDLINE, CAB Abstracts, Web of Science, and Scopus, with a publication limit of 2000-2020. All citations will be exported to EndNote® X9 Desktop and de-duplicated. The final set of articles resulting from the de-duplication will be uploaded to the Rayaan webtool as a .bib file for the first phase of the screening process (Ouzzani et al., 2016).

Search

The search strings for each one of the search databases and for each one of the transformation processes were drafted by an experienced librarian (André J. Nault) with input from experts in

the field (Drs. Kristine H. Wammer, Huan He, and William A. Arnold). The search strings can be found at the end of this protocol.

Selection of sources of evidence

The selection of relevant studies will be done with the Rayaan webtool (Ouzzani et al., 2016) by a team of four reviewers. Each article will be assessed by two independent reviewers. Reviewers will resolve any conflicts about the selection of articles into the study during weekly meetings, and with the help of a third reviewer if needed. The first screening phase will consist of only reviewing the title and abstract of each article, and the second screening phase will consist of reviewing the full text of those articles that are retained after the first screening phase. Prior to screening articles, the screening process will be pre-tested by all four reviewers with a random set of articles, and the process will be improved based on feedback from this pre-testing phase.

<u>First screening phase (title/abstract)</u>

1) Is the title/abstract available? If not, the record will be excluded.
2) Is the title/abstract in English? If not, the record will be excluded.
3) Does the title/abstract refer to primary research? If not, the record will be excluded. We will exclude reviews and conference proceedings.
4) Does the title/abstract include a natural system (river, lake, stream, creek, estuary, marine, soil, other), a laboratory experiment, or a modeling study with relevant data to the natural system of interest? If not, the record will be excluded. Engineered systems such as wastewater treatment plants will be excluded.
5) Does the title/abstract refer to marine sediments or marine biodegradation (i.e., biodegradation in marine environment)? If so, does it also refer to photolysis-

photodegradation? If the study solely reports on marine sediments and/or marine biodegradation without involving photodegradation, the study will be excluded.

6) Does the title/abstract include water, sediment, or soil samples? If not, the record will be excluded unless it is a relevant laboratory and/or modeling study.

7) Does the title/abstract evaluate any of the following processes: photolysis (UV-A/B radiation with a wavelength between 290-400 nm, sunlight, simulated sunlight, indirect photolysis induced by hydroxyl radical, carbonate radical, singlet oxygen, triplet excited organic matter), biodegradation (all systems except marine), or sorption (all systems except marine), and/or persistence? If not, the record will be excluded.

8) Does the title/abstract include any of the following target compounds: i) sulfonamides: sulfachlorpyridazine, sulfadiazine, sulfadimethoxine, sulfamethazine, sulfamethoxazole, sulfapyridine; ii) macrolides: erythromycin, roxithromycin, tylosin, azithromycin; iii) tetracyclines: chlortetracycline, doxycycline, oxytetracycline, tetracycline; iv) fluoroquinlones: ciprofloxacin, enrofloxacin, norfloxacin, ofloxacin; v) beta-lactams: penicillins (broadly); vi) others: carbadox, trimethoprim, and/or lincomycin? If not, the record will be excluded.

If the answer to any of the above questions is 'No', the record will be excluded (in the Rayyan webtool they will be labeled as 'Exclude'). If the answer to all the above questions is 'Yes' or 'Maybe' (or a combination of both), records will move to the 2nd screening phase where the full text will be reviewed (in the Rayyan webtool they will be labeled as 'Include' or 'Maybe'). The full text for included studies will be retrieved through the University of Minnesota Libraries and imported into Mendeley 1.19.8.

<u>Second screening phase (Full text)</u>

The same questions that were used during the first screening phase will be applied here. In addition, the following questions will be assessed:

1) Is the full text available? If not, the record will be excluded.

2) Does the full text include information about both a parent antibiotic compound and its product/transformation processes? If the study describes the parent compound without data for the transformation processes or products, it will be excluded.

If the answer to any of the above questions is 'No', the study will be excluded. If the answer to all the above questions is 'Yes' or 'Maybe', the study will be included.

Data charting process

Data charting will be conducted in the REDCap web application (Harris et al., 2009) independently by two reviewers. The reviewers will resolve any conflicts during weekly meetings and/or with the help of a third reviewer. The data charting tool will be pre-tested using a random sample of studies, and the tool will be improved based on the reviewer's feedback.

Data items

The data that will be extracted from each study will include the following:

1) General information: author(s), journal, publication year, aim of the study, geographic region.

2) Study characteristics: study design, system/setting used (e.g. river), sample type(s), transformation processes, time period when the study took place, specific antibiotics evaluated.

3) Outcome data: data for each transformation process/antibiotic combination will be extracted.

Critical appraisal of individual sources of evidence

A critical appraisal of individual sources of evidence will not be conducted as this is a scoping review of the literature and not a systematic review (Sargeant & O'Connor, 2020).

Synthesis of results

Studies will be grouped according to the transformation processes reported, and within those, by antibiotic compounds. Descriptive statistics will be conducted for the outcome data for each one of the transformation process/antibiotic combinations within the different study systems/settings and sample types. A narrative summary will also be conducted that will include general information and specific studies characteristics. A meta-analysis will be conducted if there are enough data for each transformation process/antibiotic compound combination. In addition, any data gaps will be identified.

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Search strings

Scopus

TITLE-ABS-KEY (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR “sulfa drug” OR macrolide OR fluoroquinolone OR beta-lactam) AND TITLE-ABS-KEY (lake OR river OR stream OR aquatic) AND TITLE-ABS-KEY (sediment) AND TITLE-ABS-KEY (adsorption OR sorption OR persistence) AND (PUBYEAR AFT 1999 AND PUBYEAR BEF 2022) AND (LIMIT-TO (LANGUAGE,"English")) AND NOT TITLE-ABS-KEY (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic OR catalytic OR Tio2 OR “titanium dioxide” OR charcoal OR bioremediation OR bioreactor OR synthesis OR “activated sludge” OR biochar)

TITLE-ABS-KEY (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR “sulfa drug” OR macrolide OR fluoroquinolone OR beta-lactam) AND TITLE-ABS-KEY (lake OR river OR stream OR aquatic OR sediment) AND TITLE-ABS-KEY (biodegradation OR biotransformation OR co-metabolism) AND (PUBYEAR AFT 1999 AND PUBYEAR BEF 2022) AND (LIMIT-TO (LANGUAGE,"English")) AND NOT TITLE-ABS-KEY (photocatalysis OR catalysis OR

catalyst OR photocatalyst OR photocatalytic OR catalytic OR Tio2 OR “titanium dioxide” OR charcoal OR bioremediation OR bioreactor OR synthesis OR “activated sludge” OR biochar)

TITLE-ABS-KEY (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR “sulfa drug” OR macrolide OR fluoroquinolone OR beta-lactam) AND TITLE-ABS-KEY (lake OR river OR stream OR aquatic OR sediment) AND TITLE-ABS-KEY (photodegradation OR phototransformation OR photolysis OR "indirect photolysis" OR "hydroxyl radical" OR "singlet oxygen") AND (PUBYEAR AFT 1999 AND PUBYEAR BEF 2022) AND (LIMIT-TO (LANGUAGE,"English")) AND NOT TITLE-ABS-KEY (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic OR catalytic OR Tio2 OR “titanium dioxide” OR charcoal OR bioremediation OR bioreactor OR synthesis OR “activated sludge” OR biochar)

CAB Abstracts

[mp=abstract, title, original title, broad terms, heading words, identifiers, cabicodes] (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR sulfa drug OR macrolide OR fluoroquinolone OR beta-lactam).mp. AND (lake OR river OR stream OR aquatic).mp. AND (sediment).mp. AND (adsorption OR sorption OR

289 persistence).mp. NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR
290 photocatalytic OR catalytic OR Tio2 OR titanium dioxide OR charcoal OR bioremediation OR
291 bioreactor OR synthesis OR activated sludge OR biochar).mp.
292
293 (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR
294 sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR
295 chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR
296 enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR
297 sulfonamide OR sulfa drug OR macrolide OR fluoroquinolone OR beta-lactam).mp. AND (lake
298 OR river OR stream OR aquatic OR sediment).mp. AND (biodegradation OR biotransformation
299 OR co-metabolism).mp. NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR
300 photocatalytic OR catalytic OR Tio2 OR titanium dioxide OR charcoal OR bioremediation OR
301 bioreactor OR synthesis OR activated sludge OR biochar).mp.
302 (sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR
303 sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR
304 chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR
305 enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR
306 sulfonamide OR sulfa drug OR macrolide OR fluoroquinolone OR beta-lactam).mp. AND (lake
307 OR river OR stream OR aquatic OR sediment).mp. AND (photodegradation OR
308 phototransformation OR photolysis OR indirect photolysis OR hydroxyl radical OR singlet
309 oxygen).mp. NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic
310 OR catalytic OR Tio2 OR titanium dioxide OR charcoal OR bioremediation OR bioreactor OR
311 synthesis OR activated sludge OR biochar).mp.

Web of Science

(ALL=((sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR "sulfa drug" OR macrolide OR fluoroquinolone OR "beta lactam") AND (lake OR river OR stream OR aquatic) AND (sediment) AND (adsorption OR sorption OR persistence) NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic OR catalytic OR Tio2 OR "titanium dioxide" OR charcoal OR bioremediation OR bioreactor OR synthesis OR "activated sludge" OR biochar))) AND LANGUAGE: (English) Indexes=SCI-EXPANDED, SSCI, A&HCI, CPCI-S, CPCI-SSH, BKCI-S, BKCI-SSH, ESCI, CCR-EXPANDED, IC Timespan=2000-2021

(ALL=((sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR “sulfa drug” OR macrolide OR fluoroquinolone OR “beta lactam”) AND (lake OR river OR stream OR aquatic OR sediment) AND (biodegradation OR biotransformation OR “co-metabolism”) NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic OR catalytic OR Tio2 OR “titanium dioxide” OR charcoal OR bioremediation OR bioreactor OR synthesis OR “activated sludge” OR biochar))) AND LANGUAGE: (English) Indexes=SCI-EXPANDED, SSCI, A&HCI, CPCI-S, CPCI-SSH, BKCI-S, BKCI-SSH, ESCI, CCR-EXPANDED, IC Timespan=2000-2021

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synthesis OR "activated sludge" OR biochar) AND 2000/01/01:2021/03/23[dp] AND (english[Language])

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(sulfachlorpyridazine OR sulfadiazine OR sulfadimethoxine OR sulfamethazine OR sulfamethoxazole OR sulfapyridine OR erythromycin OR roxithromycin OR tylosin OR chlortetracycline OR doxycycline OR oxytetracycline OR tetracycline OR ciprofloxacin OR enrofloxacin OR norfloxacin OR ofloxacin OR carbadox OR trimethoprim OR lincomycin OR sulfonamide OR “sulfa drug” OR macrolide OR fluoroquinolone OR “beta lactam”) AND (lake OR river OR stream OR aquatic OR sediment) AND (photodegradation OR phototransformation OR photolysis OR “indirect photolysis” OR “hydroxyl radical” OR “singlet oxygen”) NOT (photocatalysis OR catalysis OR catalyst OR photocatalyst OR photocatalytic OR catalytic OR

Tio2 OR “titanium dioxide” OR charcoal OR bioremediation OR bioreactor OR synthesis OR “activated sludge” OR biochar) AND 2000/01/01:2021/03/23[dp] AND (english[Language])

Total from Pubmed after de-duplication was 367.