GLOBAL CONTAMINANTS OF EMERGING CONCERN AND WASTEWATER REUSE LEACHING RISKS FOR OAHU, HAWAII

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A**bstract**

Since the 1990s, there has been mounting evidence of negative environmental impacts from persistent organic pollutants, pharmaceuticals, personal care products, endocrine-disrupting chemicals (EDC), and nanomaterials. The United States Environmental Protection Agency has coined the term "contaminants of emerging concern" (CEC) to categorize these compounds that have no regulatory standards. Studies show that CECs are detected in streams around the world and some cases in treated drinking water. The main source of CECs released into the environment is municipal wastewater effluent due to inefficient treatment removal. Of particular concern is the potential risk of Hawaii's reuse of treated wastewater for agriculture and commercial purposes, such as golf courses. The purpose of this study is to assess the leaching risks of CECs through Oahu's soils and, ultimately, to groundwater. A comprehensive literature review was conducted to identify CECs that are most frequently detected in wastewater treatment plants, have low removal efficiencies, and pose the most adverse environmental and human health risk. Eleven CECs were selected for Oahu leaching risk assessment: Carbamazepine (anticonvulsant), Propranolol (beta blocker), EDCs (Estrone, Estradiol, and Ethinylestradiol), macrolides antibiotics (Azithromycin, Clarithromycin, and Roxithromycin), sulfonamides antibiotic (Sulfamethoxazole), and quinolines antibiotics (Ciprofloxacin and Ofloxacin). To assess groundwater vulnerability from leaching contaminants, the Comprehensive Leaching Risk Assessment System model was developed by the Department of Civil and Environmental Engineering at the University of Hawaii at Manoa in partnership with the State of Hawaii Department of Agriculture and the Hawaii Department of Health - Safe Drinking Water Branch. I utilized this model to map likely, uncertain, and unlikely leaching risk of the selected CECs. The results determined that there is a leaching risk of CECs throughout the island of Oahu. This study concludes with recommendations to establish exploratory sampling of Oahu's

streams, wastewater treatment plants, and wells for the selected CECs. Future efforts should also focus on impact studies of sites currently reusing wastewater. CECs need to be taken in consideration for permitting of new wastewater reuse sites.

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List of Abbreviations

AZM	Azithromycin (macrolide antibiotic)
CAS	Conventional Activate Sludge
CBZ	Carbamazepine (anticonvulsant)
CEC	Contaminants of Emerging Concern
CIPRO	Ciprofloxacin (quinoline antibiotic)
CLR	Clarithromycin (macrolide antibiotic)
CLERS	Comprehensive Leaching Risk Assessment System
HDOH	Hawaii Department of Health
EC50	Half Maximal Effective Concentration
EDC	Endocrine Disrupting Chemicals
E1	Estrone (natural estrogen)
E2	Estradiol (natural estrogen)
EE2	Ethinylestradiol (synthetic pharmaceutical estrogen)
EPA	Environmental Protection Agency
MBR	Membrane Bioreactor
OFLOX	Ofloxacin (quinoline antibiotic)
PPCP	Pharmaceutical and Personal Care Product
PPL	Propranolol (beta blocker)
POP	Persistent Organic Pollutant
RXM	Roxithromycin (macrolide antibiotic)
SMX	Sulfamethoxazole (sulfonamides antibiotic)
USGS	United States Geological Survey
WWTP	Wastewater Treatment Plant
µg/L	Micrograms per liter
ng/L	Nanograms per liter

Introduction

Contaminants of emerging concern (CEC) is a term coined by the Environment Protection Agency (EPA) to describe compounds that are not regulated, detected in streams, and have potential negative environmental impacts. CECs consist of persistent organic pollutants, pharmaceuticals, personal care products (PPCPs), endocrine-disrupting chemicals, and nanomaterials. Common traits of CECs are low biodegradability, high environmental persistence, poorly removed in wastewater treatment plants (WWTPs), and often detected in the micro to nano grams per liter concentrations. PPCPs that are consumed by humans and livestock are excreted through urine and waste in unaltered forms. These characteristics, along with global studies, show that CECs are released into the environment from municipal wastewater effluents. CECs' leaching causes a potential risk to groundwater, which is the primary source of drinking water for the state of Hawaii and many other islands in the Pacific. On the island of Oahu, nearly all potable water is obtained from underground sources and is supplied with minimal treatment of low-level chlorination. Additionally, Hawaii is expanding wastewater reuse instead of costly disposal in the ocean but the potential of contaminating soil, water sources, and crops with a wide variety of CECs is a source of concern. The objective of this study is to evaluate global detections of CECs, select a subset to perform leaching risk assessment modeling, map likely leaching risk of the selected CECs, and make recommendations to the Hawaii Department of Health.

Methods

A literature review was performed to select CECs that were most frequently detected at WWTPs and posed the greatest potential environmental and human health risk. A risk based

selection process is necessary to prioritize the thousands of CECs in existence and in order to focus future efforts. De Voogt et al. (2009) selected seven of twenty-two criteria (such as consumption, degradability/persistence, [eco]toxicity, etc.) to rank one hundred and fifty-three CECs. Ortiz de Garcia et al. (2013) performed a quantitative study of structure–activity relationship to assess the possible adverse effects of ninety-six CECs. Zhou et al. (2013) developed a multi-step screening using criteria selection and risk quotients to rank one hundred and twenty-six CECs frequently detected in the aquatic environment. Verlicchi and Zambello (2015) evaluated one hundred and sixty-nine CECs based on the risk quotient that is the ratio between pollutant concentration and its predicted no-effect concentration. Mansour et al. (2016) performed multi-criteria decision analysis method utilizing the exposure, persistence, bioaccumulation, and toxicity 88 most commonly consumed pharmaceuticals in Lebanon. Additionally, an environmental risk assessment of predicted environmental concentrations with risk quotients was performed as an alternate method of prioritization. A literature review of sixty-two papers from twenty-four different countries were used to determine average WWTPs detection influent, effluent, removal rates, and half maximal effective concentration (EC50) for the selected CECs. All of the studied WWTPs were either membrane bioreactors (MBRs) or conventional activated sludge (CAS).

The methodological differences of multi-criteria and risk quotients makes it difficult to compare results of different studies. Criteria-based studies preform low or high rankings based on what the authors consider are the most important criteria, while risk quotients vary due to different methods of predicting estimated concentrations and predicted no-effect concentrations. Other variables tend to change based on location, such as, prescription rates, availability of sales data, and population density per WWTPs. Despite these differences, anticonvulsants, antibiotics,

estrogen hormones, and beta blockers are frequently detected and identified as hazards to the aquatic environments. Therefore, I selected eleven CECs: carbamazepine (anticonvulsant), EDCs (estrone, estradiol, and ethinylestradiol), macrolides antibiotics (azithromycin, clarithromycin, and roxithromycin), sulfonamides antibiotic (sulfamethoxazole), quinolines antibiotics (ciprofloxacin and ofloxacin), and propranolol (beta blocker) (Table 2).

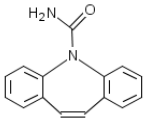
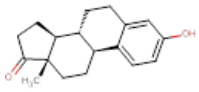
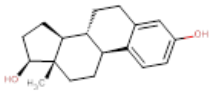
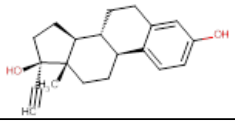
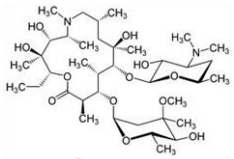
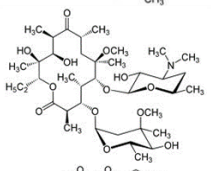
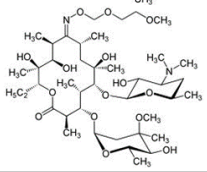
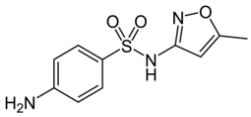
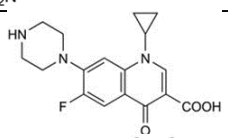
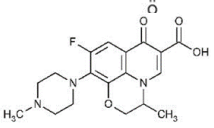
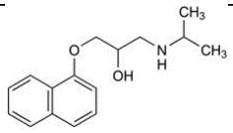
Selected CECs	CAS Number	Molecular Structure	MW (g/mol)	pKa	Log K_{ow}	S_w 25°C (mg/L)	Log K_d
Carbamazepine (CBZ)	298-46-4		236.28	13.9	2.45	17.66	0.10
Estrone (E1)	53-16-7		270.366	10.25	3.43	146.80	2.4-2.9
Estradiol (E2)	50-28-2		272.38	10.27	3.94	81.97	2.4-2.8
Ethinylestradiol (EE2)	57-63-6		296.403	10.24	4.12	116.40	1.2-8
Azithromycin (AZM)	83905-01-5		748.984	8.7	4.02	0.06	2.5-2.7
Clarithromycin (CLR)	81103-11-9		747.953	8.99	3.16	0.34	0.4-1.7
Roxithromycin (RXM)	80214-83-1		837.047	8.8	2.75	0.02	0.2-0.3
Sulfamethoxazole (SMX)	723-46-6		253.28	5.7	0.89	3942	2.1-2.7
Ciprofloxacin (CIPRO)	85721-33-1		331.346	6.38	0.4	11480	4.3
Ofloxacin (OFLOX)	82419-36-1		361.368	5.97	0.35	28260	4.2
Propranolol (PPL)	525-66-6		259.34	9.42	3.48	288.00	2.6

Table 2. Summary of Selected CEC (values obtained from Verlicchi and Zambello 2015).

In order to economically evaluate the leaching potential of the selected CECs, I utilized the Comprehensive Leaching Risk Assessment System (CLERS) model, which was recently developed by the Department of Civil and Environmental Engineering at the University of Hawaii based on the original model of Stenemo et al. (2007) that is used by the Hawaii Department of Agriculture as part of the first-tier screening tool for pesticides in Hawaii. Due to the growing concern of CECs, the Hawaii Department of Health needed an assessment tool to evaluate groundwater vulnerability and CLERS was expanded to include liquid-vapor partitioning for the retardation factor and vapor loss at the soil surface for the attenuation factor (Ray et al. 2014). In this study, CLERS Version 3 python script was used with ArcGIS 10.1 to classify the selected CECs as likely, uncertain, or unlikely to leach based on chemical and soil properties. The model assesses the risk by comparing the chemical's calculated attenuation factor against those of two Hawaii-known reference pesticides: atrazine (leacher), and endosulfan (non-leacher). CLERS shows good agreement when tested against the numerical model HYDRUS-1D (Šimůnek et al. 2013) that describes the complex movement of chemicals in Hawaii (Ray et al. 2014). Additionally, an updated recharge model for Oahu (Engott et al. 2015) was used instead of the original model (Rotzoll and El-Kadi 2007) embedded in the CLERS Version 3 model. The 2015 values more accurately represent average recharge for Oahu as shown in Figure 1.

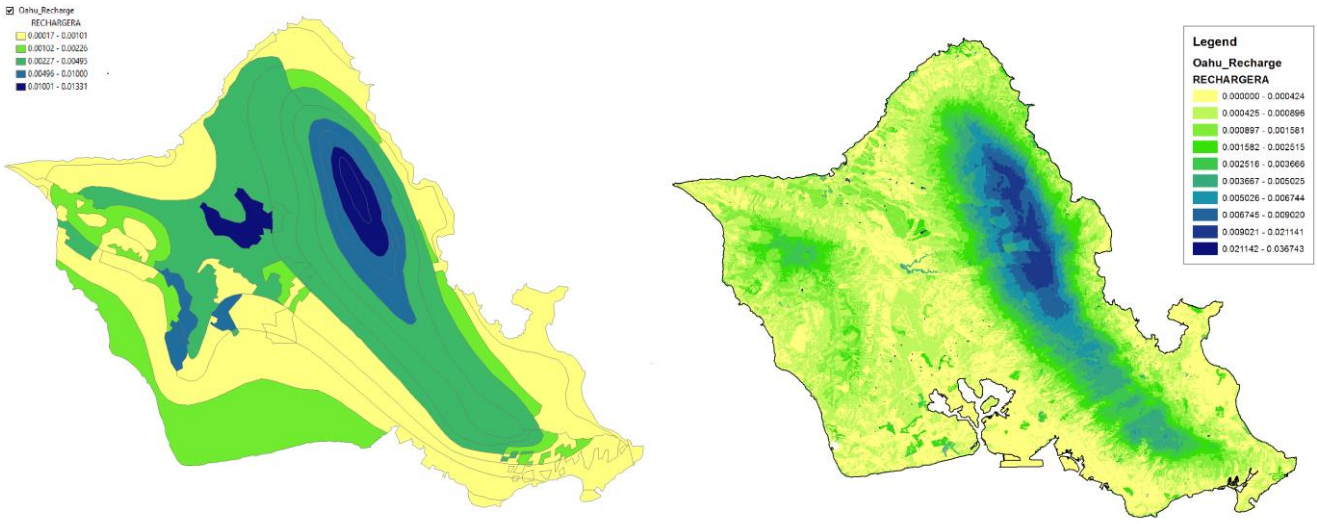


Figure 1. Left – Rotzoll and El-Kadi 2007 Recharge, Right – Engott et al. 2015 Recharge

There are two input parameters needed for the calculation of the attenuation factor in the CLERS model: the log of soil organic carbon sorption coefficient (K_{oc}) and the soil half-life ($t_{1/2}$) of the chemical compound. However, K_{oc} of the selected CECs is poorly understood. As a first approximation, I used the Estimation Program Interface (EPI) Suite version 4.1 (US EPA 2012), specifically, the KOCWIN sub-program to estimate the selected CECs K_{oc}. A literature review was conducted to identify soil half-lives and the EPI Suite was used to estimate unknown values (Table 2).

Selected CECs	Log K_{oc}	SD_K_{oc}	$t_{1/2}$ (d)	SD $t_{1/2}$ (d)	$t_{1/2}$ Reference
Carbamazepine (CBZ)	2.23*	0	495	36	Walters et al. 2010
Estrone (E1)	3.019*	0	2.7	0	Xuan et al. 2008
Estradiol (E2)	2.899*	0	0.92	0	Xuan et al. 2008
Ethinylestradiol (EE2)	2.71*	0	1.9	0	Xuan et al. 2008
Azithromycin (AZM)	1.676*	0	770	181	Walters et al. 2010
Clarithromycin (CLR)	1.371*	0	360*	0	*
Roxithromycin (RXM)	0.858*	0	360*	0	*
Sulfamethoxazole (SMX)	1.536*	0	23.4	42	Kodesova et al. 2015
Ciprofloxacin (CIPRO)	-0.004*	0	2310	1155	Walters et al. 2010
Ofloxacin (OFLOX)	-0.004*	0	1386	434	Walters et al. 2010
Propranolol (PPL)	2.451*	0	620	0	Yamamoto et al. 2008

*Estimated values by EPI Suite (US EPA 2012)

Table 2. Summary of Log Koc and $t_{1/2}$ for the selected CECs

Selected Contaminants of Emerging Concern and Impacts

CBZ is an anticonvulsant and mood stabilizing drug used primarily in the treatment of epilepsy and bipolar disorder. Although the impacts are not well understood, CBZ is lipophilic and has a low biodegradability, which allows it to persist in the environment. The drug has average removal efficiencies at WWTPs below 10% (Zhang et al. 2008). CBZ was found to be detrimental to mussels (Mytilus galloprovincialis) by acting on specific biochemical pathways for which it was designed and that are evolutionally conserved (Martin-Diaz et al. 2009). A study

on aquatic insects, Chironomus riparius, showed CBZ caused a blockade of pupation and emergence of nonbiting midge (Oetken et al. 2005).

E1 and E2 are endogenous estrogens while EE2 is a derivative of estradiol used in birth control and hormone therapy. Human-excreted estrogens are poorly treated in WWTPs, septic tanks, and cesspools and therefore, are released into the environment. Estrogenic hormones disrupt the endocrine system functions of animals and humans, which produce adverse reproductive, neurological, and developmental effects. Of particular concern, E1, E2, and EE2 exert physiological effects at very low concentrations relative to other EDCs. Exposure to estrogen levels as low as 1 ng/L have caused feminization of male trout (Hansen et al. 1998) and intersex of roach (Rutilus rutilus) gonads (Lange et al. 2008). In the South Branch of the Potomac River and select nearby drainages, more than 80% of all the male smallmouth bass sampled had oocytes (immature egg cells) growing within their testicular tissue (Blazer et al. 2007). Studies from a wide range of rivers in the British Isles demonstrated that intersexuality of roaches is occurring at rates up to 100%, downstream of WWTPs verse controls at 4% (Jobling et al. 1998). Recent studies of trout fry exposed to EE2 significantly impaired their growth rate and caused intersex of males as compared to controls (Depiereux et al. 2014).

The three most studied and detected types of antibiotics are macrolides, sulfonamides, and quinolines. In 2010, over 258 million prescriptions of antibiotics were made in the United States and macrolides were the most common with AZM being the most prescribed antibiotic (Hicks and Taylor 2013). Macrolides work by targeting protein synthesis through the ribosomal subunits, sulfonamides work by inhibiting the enzyme dihydropteroate, and quinolones work by interfering with the nucleic acid synthesis (Sengupta et al. 2013).

Antibiotics are a potential environmental pollutant because they are developed with the intention of having a biological effect, are metabolically stable, widely prescribed, and poorly removed at WWTPs. In 2003, the Institute of Medicine identified antibiotic resistance as a key microbial threat to health and it is estimated that 50% of antibiotic prescriptions may be unnecessary (Centers for Disease Control and Prevention 2011). While antibiotics are generally associated with human medicine, they are also substantially used as growth promoters, feed additives in livestock production, and in veterinary medicine. Antibiotic use, especially when excessive or unnecessary, combined with reuse of contaminated wastewaters will increase the chance of antibiotic-resistance pathogens. A recent study proved that co-aggregates of bacteria (such as biofilms) are ideal environments for fast adaptations to antibiotics present in aquatic systems (Corno et al. 2014). Additionally, bacterial infections continue to be one of the leading causes of morbidity and mortality worldwide, attributed in part to the evolution and dissemination of antibiotic resistance genes (D'costa et al. 2007).

PPL is a β blocker used in the treatment of diverse diseases including high blood pressure, irregular heart rate, anxiety, and migraines. A literature review of PPL's toxicity to aquatic organisms has a number of mixed results. PPL has significant effects on growth, heart rate, and abdominal appendage movement on water fleas (Daphnia magna) (Jeong et al. 2015). Another study showed that 0.5 µg/L of PPL to Japanese rice fish was enough to significantly decrease the number of eggs produced and the number of viable eggs that hatched (Huggett et al. 2002). On the other hand, studies of rainbow trout required much higher concentrations (10 mg/L) to affect reproduction (Owen et al. 2009). Another study of fathead minnows (Pimephales promelas) shows adverse reproduction effects at concentrations of 1 mg/L (Giltrow et al 2009).

While those studies showed a negative impact, they were at concentrations much higher than environmentally relevant.

The synergist effects of multiple CECs have been rarely studied but are often mentioned as a potential concern. Estrogenic compounds, when present in mixtures, add incrementally to the total estrogenic effects, even when each component is present at concentrations that individually produce no detectable effects (Payne et al. 2000, Rajapakse et al. 2002). A study on the effect of coexistent sulfonamide antibiotics with E2 significantly reduced the degradation rate (from 0.750 to 0.264 per day) and extended half-life (from 0.92 to 2.6 days) of E2 (Xuan et al. 2008).

Results

The data and referenced studies used for calculations documented in this section are detailed in Appendix A and summarized in Table 3. The CLERS classification maps for the selected CECs are displayed in Appendix B and summarized in Table 4. A map of tax map keys that are currently reusing wastewater in Oahu is located in Appendix C.

Carbamazepine	Min	Max	Avg
Influent (µg/L)	0.00	21.5	**2.87**
Effluent (µg/L)	0.00	19.8	**2.37**
Removal Efficiencies	-47	97	**7**

Roxithromycin	Min	Max	Avg
Influent (µg/L)	0.01	17.00	**2.48**
Effluent (µg/L)	0.01	5.00	**0.59**
Removal Efficiencies	-80	75	**26**

Estrone	Min	Max	Avg
Influent (µg/L)	0.020	0.070	**0.035**
Effluent (µg/L)	0.002	0.072	**0.017**
Removal Efficiencies	-83	112	**59**

Sulfamethoxazole	Min	Max	Avg
Influent (µg/L)	0.00	10.00	**1.22**
Effluent (µg/L)	0.00	5.00	**0.43**
Removal Efficiencies	-120	100	**42**

Estradiol	Min	Max	Avg
Influent (µg/L)	0.003	3.00	**0.19**
Effluent (µg/L)	0.00	0.054	**0.006**
Removal Efficiencies	21.8	99.9	**78**

Ciprofloxacin	Min	Max	Avg
Influent (µg/L)	0.09	13.60	**1.37**
Effluent (µg/L)	0.01	2.37	**0.55**
Removal Efficiencies	-44	96	**60**

Ethinylestradiol	Min	Max	Avg
Influent (µg/L)	0.00	0.07	**0.02**
Effluent (µg/L)	0.00	0.010	**0.0017**
Removal Efficiencies	25	94	**70**

Ofloxacin	Min	Max	Avg
Influent (µg/L)	0.02	31.70	**3.32**
Effluent (µg/L)	0.02	0.86	**0.28**
Removal Efficiencies	13	99	**64**

Azithromycin	Min	Max	Avg
Influent (µg/L)	0.04	1.34	**0.36**
Effluent (µg/L)	0.038	0.060	**0.0495**
Removal Efficiencies	5	74.3	**38**

Propranolol	Min	Max	Avg
Influent (µg/L)	0.01	1.90	**0.25**
Effluent (µg/L)	0.00	0.56	**0.18**
Removal Efficiencies	0	96	**46**

Clarithromycin	Min	Max	Avg
Influent (µg/L)	0.03	4.82	**1.31**
Effluent (µg/L)	0.15	0.46	**0.29**
Removal Efficiencies	0	92	**46**

Table 3. Summary of WWTP influent, effluent, and removal efficiencies of selected CECs.

Selected CECs	CLERS Classification
Carbamazepine (CBZ)	Likely
Estrone (E1)	Unlikely
Estradiol (E2)	Unlikely
Ethinylestradiol (EE2)	Unlikely
Azithromycin (AZM)	Likely*
Clarithromycin (CLR)	Likely
Roxithromycin (RXM)	Likely*
Sulfamethoxazole (SMX)	Uncertain - Likely
Ciprofloxacin (CIPRO)	Likely
Ofloxacin (OFLOX)	Likely*
Propranolol (PPL)	Likely

Table 4. Summary of CLERs leaching classification of selected CECs. * notes some coastal areas are classified as unlikely while the majority of the map is classified as likely.

Discussion

The selected CECs do not imply an all-inclusive list but represent the most frequently identified in the above risk assessment studies, the greatest availability of WWTP sampling data, and having highest potential environmental impacts. While there are other frequently detected CECs, such as caffeine, Ibuprofen, salicylic acid, DEET, and acetaminophen, they pose a low environmental risk and tend to be used as human tracers. Other CECs that have been frequently detected but have poorly studied impacts are: aciclovir (anti-infective), bisacodyl (alimentary tract and metabolismconstipation), clonixin (musculo-skeletal), diclofenac (anti-inflammatory), dimenhydrinate (respiratory), domperidone (alimentary tract and metabolism-gastronomical), naftidrofuryl (cardiovascular), naproxen (anti-inflammatory), sertraline (serotonin reuptake

inhibitor), and fragrances (galaxolide, tonalid, phantolide). The β-lactam antibiotics (amoxicillin, cephalexin, penicillin) could have been selected, but there are limited WWTP sampling data. Lastly, expansion of the selected CECs could include anticonvulsant nifedipine, beta blockers metoprolol and atenolol, antibiotics norfloxacin and erythromycin, and EDCs related to testosterone. The above CECs are worth considering for future studies as environmental impacts and sampling data become more available.

Potential reasons for prolonged detection of CECs may include strong electrostatic sorption to biosolids and chemical aging which results in low bioavailability (Walters et al. 2010). The aging mechanisms relate to the migration of molecules into very small (<100 nm) nanopores sites within the soil matrix where the chemicals become lodged and microorganisms are unable to access (Alexander 2000). The main sorbent for hydrophobic molecules in soils is organic matter; therefore, CECs may be tightly bound in soil nanopores which leads to extended residence times (Alexander 2000). The electrostatic sorption and aging mechanism may explain why soil $t_{1/2}$ of antibiotics range 770-2300 days while the EPI Suite estimates 360 days for most antibiotics.

There have been limited studies of the selected CECs in Hawaii but CBZ and SMX were detected in treated wastewater effluent and at offsite wells and springs at Kealakehe, Hawaii (Hunt, 2008). CBZ and SMX were also detected in a multitracer study which was a strong indication of treated effluent presence within the modeled plume footprint at Kihei and at the Submarine Springs (Hunt and Rosa, 2009). Studies of estrogen compounds in Hawaii soils showed limited mobility in the top soil with the greatest risk from cesspools or septic systems close to groundwater (D'Alessio et al. 2014). A 2015 USGS and Honolulu Board of Water Supply survey to evaluate the potential effects of irrigation with treated wastewater on

groundwater quality in North-Central Oahu found CBZ, SMZ, and PPL (values included in Table 5) in effluent and an irrigation ditch among 51 other CECs.

Selected CECs	Effluent (µg/L)	PNEC (µg/L)	Based On	Reference	Wahiawa WWTP Effluent (µg/L)	Kaukonahua Ditch (µg/L)	Haleiwa P2 (µg/L)
Carbamazepine (CBZ)	2.37	13.8	Water Flea	Ferrari et al., 2004	0.194	0.022	0.010
Estrone (E1)	0.017	0.006	Fish	Caldwell et al., 2012			
Estradiol (E2)	0.0058	0.002	Fish	Caldwell et al., 2012			
Ethinylestradiol (EE2)	0.0017	0.0001	Fish	Caldwell et al., 2012			
Macrolides							
Azithromycin (AZM)	0.0495	0.15	Algea	Kümmerer et al., 2003			
Clarithromycin (CLR)	0.29	0.07	Algea	Boillot et al., 2008			
Roxithromycin (RXM)	0.59	4	Algea	Sanderson et al., 2003			
Sulfonamides							
Sulfamethoxazole (SMX)	0.43	0.027	Algea	Sanderson et al., 2003	0.316	0.130	ND
Quinolines							
Ciprofloxacin (CIPRO)	0.55	938	Algea	Sanderson et al., 2003			
Ofloxacin (OFLOX)	0.28	0.016	Algea	Ferrari et al., 2004			
Propranolol (PPL)	0.18	0.224	Diatoms	Ferrari et al., 2004	0.042	0.012	ND

Table 5. Summary of average global effluent, predicted no-effect concentrations, and Oahu detections.

If we consider the average global effluents from WWTPs and predicted no-effect concentrations (PNEC) (Table 5) for the selected CECs, E1, E2, EE3, CLR, SMX, and OFLOX average effluents are above PNEC (colored red) indicating higher potential risk than CBZ, AZM, RXM, CIPRO, and PPL which are lower than PNEC (colored green). The values obtained from the 2015 USGS survey in Oahu echo the global risk; SMX sampled concentration is above PNEC and CBZ and PPL sampled concentrations are below PNEC. The blank areas indicate the CEC was not tested for and ND equals not detected.

Conclusions

The global literature review shows that WWTPs do not effectively treat the selected CECs. Additionally, E1, E2, EE3, CLR, SMX, and OFLOX have average effluents greater than the PNEC values implying higher risk potential for those compounds. CLERS modeling classified the selected EDCs as unlikely leachers, SMX as uncertain to likely leacher, and the remaining selected CECs as likely leachers. The reuse of wastewater for agricultural and commercial purposes should be critically reviewed. The Hawaii Department of Health needs to perform exploratory sampling of streams, wells, and WWTP effluents to establish an understanding of what CECs are present and their concentrations. At sites where recycled water use on soils is practiced, monitoring systems must be in place to detect and track the presence of CECs. Hawaii needs to be proactive and contribute its knowledge to this global issue of CECs and ultimately ensure the long-term safety of our drinking water and environment.

Appendix A – Selected CECs and WWTP influents, effluents, and removal efficiencies.

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Carbamazepine	1.68	Bendz et al. 2005	0.3-1	Andreozzi et al., 2003;	30	Bendz et al., 2005;
	0.7*/0.32-0.7	Clara et al., 2005a,	1.18	Bendz et al., 2005;	7/13*	Bernhard et al., 2006;
	0.32-1.2	Clara et al., 2005b;	0.794*/0.465-0.952	Clara et al., 2005a,	0	Castiglioni et al., 2006;
	0.3	Conti et al., 2011;	1.147*/0.465-1.337	Clara et al., 2005b;	0/0*	Clara et al., 2004,
	<0.2-0.59	Foster, 2007;	1.519	Coetsier et al 2009;	-47-(-3)/-13*	Clara et al., 2005a,
	0.12-0.31	Gómez et al., 2007;	<0.05-0.15	Foster, 2007;	-43-(-3)/4.4*	Clara et al., 2005b;
	0.1-3.11	Kasprzyk-Hordern et al.,	0.11-0.23	Gómez et al., 2007;	75	Foster, 2007;
	0.5	Khan and Ongerth, 2005;	0.15-2.32	Kasprzyk-Hordern et	13	Gómez et al., 2007;
	<0.005-0.45	Choi et al., 2008;	0.5	Khan and Ongerth,	13	Kasprzyk-Hordern et al.,
	0.015-0.27	Nakada et al., 2006;	<0.005-0.195	Choi et al., 2008;	0	Khan and Ongerth,
	1.3-2	Paxéus, 2004;	0.073-0.729	Kim et al., 2007;	30-64	Choi et al., 2008;
	0.054-0.22	Radjenovic et al., 2009;	0.14-0.26	Muñoz et al., 2009;	14/35/11*	Kreuzinger et al., 2004;
	19.5*	Reif et al., 2008;	0.011-0.16	Nakada et al., 2006;	-122-77.6	Nakada et al., 2006;
	0.1-0.17	Rosal et al., 2010;	0.1-1.2	Paxéus, 2004;	10-53	Paxéus, 2004;
	<loq/2.15	Santos et al., 2007,	17.8*	Reif et al., 2008;	5	Radjenovic et al., 2007,
	<loq-3.78	Santos et al., 2009;	0.069-0.173	Rosal et al., 2010;	5	Radjenovic et al., 2009;
	0.2*	Snyder et al., 2006;	<loq-1.29	Santos et al., 2007,	9*	Reif et al., 2008;
	21.5	Suárez et al., 2005;	<loq-1.29	Santos et al., 2009;	9.5	Rosal et al., 2010;
	1	Wick et al, 2009	<0.01*	Snyder et al., 2006;	-67-(-4)	Santos et al., 2007,
			19.8	Suárez et al., 2005;	7-11	Santos et al., 2009;
			2.1-6.3	Ternes, 1998;	97*	Snyder et al., 2006;
			2.1	Ternes et al., 2003;	7.9	Suárez et al., 2005;
			0.74-0.92	Wick et al, 2009	7	Ternes, 1998;
					-44	Vieno et al., 2007;
					-12	Wick et al, 2009

loq = limit of quantification = reporting limit
WWTP Types: * = MBRs otherwise CAS

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Estrone	0.05-0.07	Andersen et al., 2003;	0.0005	Andersen et al., 2003;	99.24	Andersen et al., 2003;
	0.03-0.07	Baronti et al., 2000;	0.005-0.044	Baronti et al., 2000;	12-92.53	Baronti et al., 2000;
	0.002	Carballa et al., 2004,	<loq-0.0044	Carballa et al., 2004,	-83	Carballa et al., 2004,
	0.071*/0.034-0.67	Clara et al., 2005a;	<loq-0.072/0.002*	Clara et al., 2005a;	-40	Carballa et al., 2005;
	0.025*/0.032	Joss et al., 2005;	0.002/0.002*	Joss et al., 2005;	112-99.93/97.18*	Clara et al., 2005a;
	0.03	Lishman et al., 2006;	0.002-0.036	Kim et al., 2007;	96/96*	Joss et al., 2005;
	0.02-0.19	Nakada et al., 2006;	0.0076-0.038	Lishman et al., 2006;	57	Lishman et al., 2006;
	0.014	Zorita et al., 2009	0.0028-0.11	Nakada et al., 2006;	83.9-90.3	Nakada et al., 2006;
			0.07	Zorita et al., 2009	83	Ternes et al., 1999

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Estradiol	0.012-0.02	Andersen et al., 2003;	<0.001	Andersen et al., 2003;	75-92.22	Baronti et al., 2000;
	0.008-0.016	Baronti et al., 2000;	0.0007-0.002	Baronti et al., 2000;	46	Carballa et al., 2005;
	0.035-0.067/0.067*	Clara et al., 2005a	<loq	Carballa et al., 2004,	98	Foster, 2007;
	<0.08-3	Foster, 2007;	<loq-0.03/<loq*	Clara et al., 2005a;	98/99*	Joss et al., 2005;
	0.04*/0.003	Joss et al., 2005;	<0.02-0.054	Foster, 2007;	75	Lishman et al., 2006;
	0.01	Lishman et al., 2006;	0.0002	Joss et al., 2005;	99.9	Ternes et al., 1999a;
	0.003	Zorita et al., 2009	<0.001	Kim et al., 2007;	21.8	Zorita et al., 2009
			<loq	Lishman et al., 2006;		
			0.0025	Zorita et al., 2009		

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Ethinylestradiol	0.002-0.004	Baronti et al., 2000;	0.0004-0.0008	Baronti et al., 2000;	60-86.66	Baronti et al., 2000;
	0.004-0.07/0.02*	Clara et al., 2005a	<loq-0.005/0.004*	Clara et al., 2005a;	70/70*	Clara et al., 2004,
	0.04	Foster, 2007;	<0.02-0.01	Foster, 2007;	75	Foster, 2007;
	0.002	Joss et al., 2005;	0.0002/0.0002*	Joss et al., 2005;	94/76*	Joss et al., 2005;
			0.0013	Kim et al., 2007;	70-81/25*-66*	Kreuzinger et al., 2004;
			<loq	Zorita et al., 2009	78	Ternes et al., 1999

loq = limit of quantification = reporting limit
WWTP Types: * = MBRs otherwise CAS

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Azithromycin	0.16-1.34	Ghosh et al., 2009;	0.04-0.38	Göbel et al., 2005,	45/39	Ghosh et al., 2009;
	0.09-0.38	Göbel et al., 2005,	0.06	Yasojima et al., 2006	18	Göbel et al., 2005,
	0.26	Yasojima et al., 2006			5*-24*	Göbel et al., 2007;
					74.3	Yasojima et al., 2006

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Clarithromycin	1.129-4.82	Ghosh et al., 2009;	0.15-0.46	Göbel et al., 2005,	0	Castiglioni et al., 2006;
	0.33-0.6	Göbel et al., 2005,	0.21	Ternes et al., 2003;	50-83	Ghosh et al., 2009;
	0.647	Yasojima et al., 2006	0.35	Yasojima et al., 2006	32	Göbel et al., 2005,
					4.5/41*-88*	Göbel et al., 2007;
					62/92*	Sahar et al., 2011;
					45.9	Yasojima et al., 2006

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Roxithromycin	0.025-0.078	Clara et al., 2005b;	0.042*/0.045/0.057/0.036	Clara et al., 2005b;	-80-43.8/34.4*	Clara et al., 2005b;
	0.096-0.209	Ghosh et al., 2009;	0.01-0.03	Göbel et al., 2005,	-71	Ghosh et al., 2009;
	0.01-0.04	Göbel et al., 2005,	5*	Reif et al., 2008;	0	Göbel et al., 2005,
	17*	Reif et al., 2008;	0.05	Ruel et al., 2010;	19/39*-62*	Göbel et al., 2007;
	0.08	Ruel et al., 2010;	0.54	Ternes et al., 2003,	-4-61/75*	Kreuzinger et al., 2004;
	0.018	Watkinson et al., 2007;	0.1	Watkinson et al., 2007;	40-46	Li and Zhang, 2011
	0.04	Xu et al., 2007	0.035	Xu et al., 2007	71*	Reif et al., 2008;
					37.5	Ruel et al., 2010;
					22/59*	Sahar et al., 2011;
					12.5	Xu et al., 2007

loq = limit of quantification = reporting limit
WWTP Types: * = MBRs otherwise CAS

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Sulfamethoxazole	0.02	Bendz et al., 2005;	0.01-0.09	Andreozzi et al., 2003;	21	Brown et al., 2006;
	0.39	Brown et al., 2006;	0.07	Bendz et al., 2005;	57	Carballa et al., 2004,
	<loq-0.58	Carballa et al., 2004,	0.31	Brown et al., 2006;	46	Carballa et al., 2005;
	0.15-0.98	Choi et al., 2008;	0.25	Carballa et al., 2004,	24	Castiglioni et al., 2006;
	0.02-0.075	Clara et al., 2005b;	0.025-0.5	Choi et al., 2008;	41-80	Choi et al., 2008;
	<0.2	Foster, 2007;	0.05-0.09/<loq*	Clara et al., 2005b;	32	Clara et al., 2005b;
	0.23-0.57	Göbel et al., 2005,	0.025	Foster, 2007;	75	Foster, 2007;
	0.17-1.25	Karthikeyan and Meyer, 2006;	0.13-0.84	Göbel et al., 2005,	54-71	García-Galán et al., 2011
	0.02-0.27	Kasprzyk-Hordern et al., 2009;	0.05-0.21	Karthikeyan and Meyer, 2006;	26-39	Ghosh et al., 2009;
	0.14-0.23	Lindberg et al., 2005;	0.004-0.044	Kasprzyk-Hordern et al., 2009;	35	Göbel et al., 2005,
	5.45-7.91	Peng et al., 2006;	0.003-0.4	Kim et al., 2007;	4.5/37*-38*	Göbel et al., 2007;
	0.25-1.3	Radjenovic et al., 2009;	0.13	Lindberg et al., 2005;	-120	Karthikeyan and Meyer, 2006;
	10*	Reif et al., 2008;	0.18	Muñoz et al., 2009;	83	Kasprzyk-Hordern et al., 2009;
	0.16-0.53	Rosal et al., 2010;	<loq	Peng et al., 2006;	62/57*	Kreuzinger et al., 2004;
	0.53	Ruel et al., 2010;	5*	Reif et al., 2008;	62-90	Li and Zhang, 2011
	1.11*	Snyder et al., 2006;	0.1-0.3	Rosal et al., 2010;	42-100	Lindberg et al., 2005;
	0.36	Watkinson et al., 2007;	0.3	Ruel et al., 2010;	99	Peng et al., 2006;
	0.01	Xu et al., 2007	<0.01*	Snyder et al., 2006;	55.6/60.5*	Radjenovic et al., 2007,
			0.62	Ternes et al., 2003;	73.8/78.3*-80.8*	Radjenovic et al., 2009;
			0.27	Watkinson et al., 2007	50*	Reif et al., 2008;
					17.3	Rosal et al., 2010;
					41.5	Ruel et al., 2010;
					10/0*	Sahar et al., 2011;
					100*	Snyder et al., 2006;
					25	Watkinson et al., 2007;
					-20	Xu et al., 2007

loq = limit of quantification = reporting limit
WWTP Types: * = MBRs otherwise CAS

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Ciprofloxacin	0.09	Costanzo et al., 2005;	0.04-0.07	Andreozzi et al., 2003;	73*	Baumgarten et al., 2007
	0.231-0.195	Ghosh et al., 2009;	0.13	Costanzo et al., 2005;	63	Castiglioni et al., 2006;
	0.315-0.57	Golet et al., 2003;	0.079-0.1	Golet et al., 2003;	-44	Costanzo et al., 2005;
	0.21	Karthikeyan and Meyer, 2006;	0.06	Karthikeyan and Meyer, 2006;	50-73	Ghosh et al., 2009;
	0.09-0.194	Lindberg et al., 2005,	0.007-0.032	Lindberg et al., 2005,	78	Golet et al., 2003;
	0.21-0.228	Lindberg et al., 2006;	0.03-0.05	Lindberg et al., 2006;	71.43	Karthikeyan and Meyer, 2006;
	0.16-13.6	Rosal et al., 2010;	2	Muñoz et al., 2009;	18/55	Li and Zhang, 2011;
	3.8	Watkinson et al., 2007;	2.37	Rosal et al., 2010;	72-96	Lindberg et al., 2005,
	0.32	Zorita et al., 2009	0.64	Watkinson et al., 2007;	79	Lindberg et al., 2006;
			0.094	Zorita et al., 2009	57	Rosal et al., 2010;
					86	Vieno et al., 2007;
					83	Watkinson et al., 2007;
					71	Zorita et al., 2009

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Ofloxacin	0.47	Brown et al., 2006;	0.31-0.58	Andreozzi et al., 2003;	77	Brown et al., 2006;
	0.287	Lindberg et al., 2005;	0.11	Brown et al., 2006;	57	Castiglioni et al., 2006;
	0.52-5.56	Peng et al., 2006;	0.045	Lindberg et al., 2005;	26-59	Li and Zhang, 2011
	0.89-31.7	Radjenovic et al., 2009;	0.04-0.86	Peng et al., 2006;	84	Lindberg et al., 2005;
	0.84-5.29	Rosal et al., 2010;	0.81	Rosal et al., 2010;	85-99	Peng et al., 2006;
	0.077	Xu et al., 2007;	0.048	Xu et al., 2007;	23.8/94*	Radjenovic et al., 2007,
	0.022	Zorita et al., 2009	0.019	Zorita et al., 2009	75.8/91.3*-95.2*	Radjenovic et al., 2009;
					64	Rosal et al., 2010;
					83	Vieno et al., 2007;
					37.66	Xu et al., 2007;
					13	Zorita et al., 2009

loq = limit of quantification = reporting limit
WWTP Types: * = MBRs otherwise CAS

Contaminant	Influent (ug/L)		Effluent (ug/L)		Removal Efficiencies (%)	
Propranolol	0.05	Alder et al., 2010;	0.03	Alder et al., 2010;	33	Alder et al., 2010;
	0.05	Bendz et al., 2005;	0.01-0.09	Andreozzi et al., 2003;	59	Kasprzyk-Hordern et al., 2009;
	0.11-1.9	Kasprzyk-Hordern et al., 2009;	0.03	Bendz et al., 2005;	28.48-34.69	Maurer et al., 2007;
	0.05-0.17	Maurer et al., 2007;	0.56	Coetsier et al., 2009;	58.8/65.5*-77.6*	Radjenovic et al., 2009;
	0.1-1.13	Radjenovic et al., 2009;	0.13-0.523	Kasprzyk-Hordern et al., 2009;	1	Rosal et al., 2010;
	0.08	Roberts and Thomas, 2006;	0.032-0.123	Maurer et al., 2007;	96	Ternes, 1998;
	0.012-0.06	Rosal et al., 2010;	0.39	Roberts and Thomas, 2006;	0	Wick et al., 2009
	0.073	Wick et al., 2009	<loq-0.057	Rosal et al., 2010;		
			0.17-0.29	Ternes, 1998;		
			0.18	Ternes et al., 2003;		
			0.058	Wick et al., 2009		

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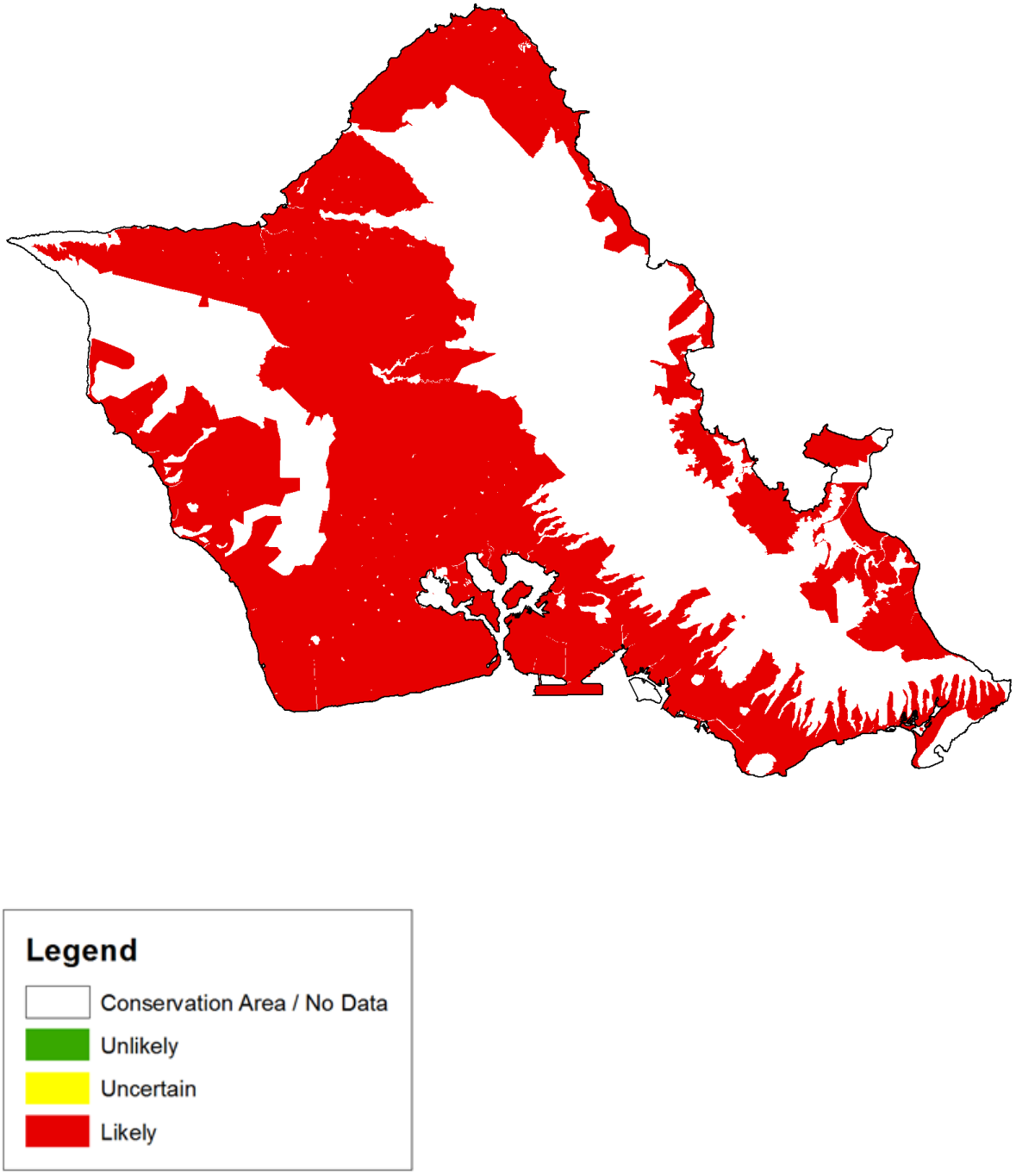
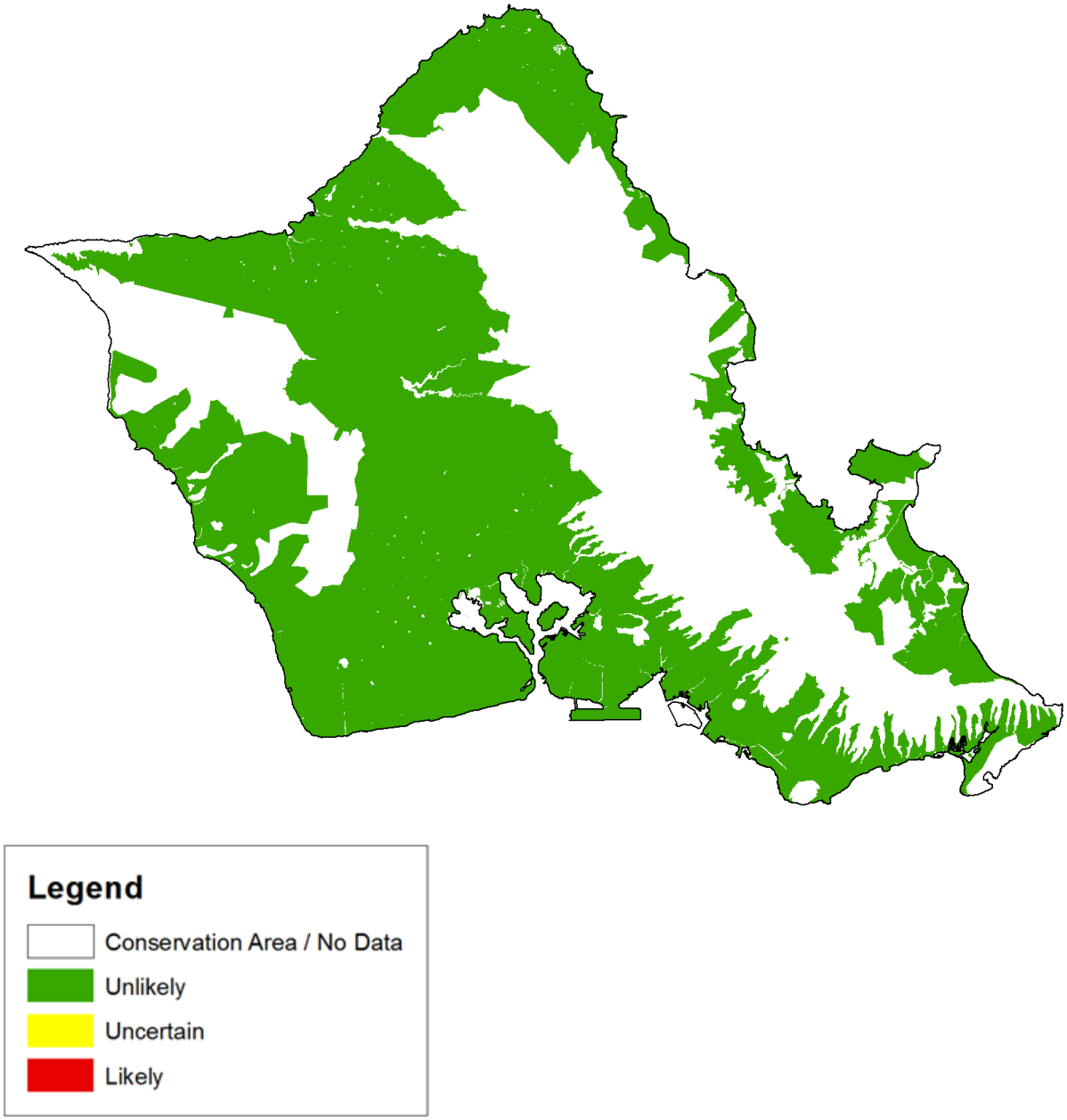
WWTP Types: * = MBRs otherwise CAS

Appendix B – CLERS Leaching Classification Maps for Oahu, Hawaii

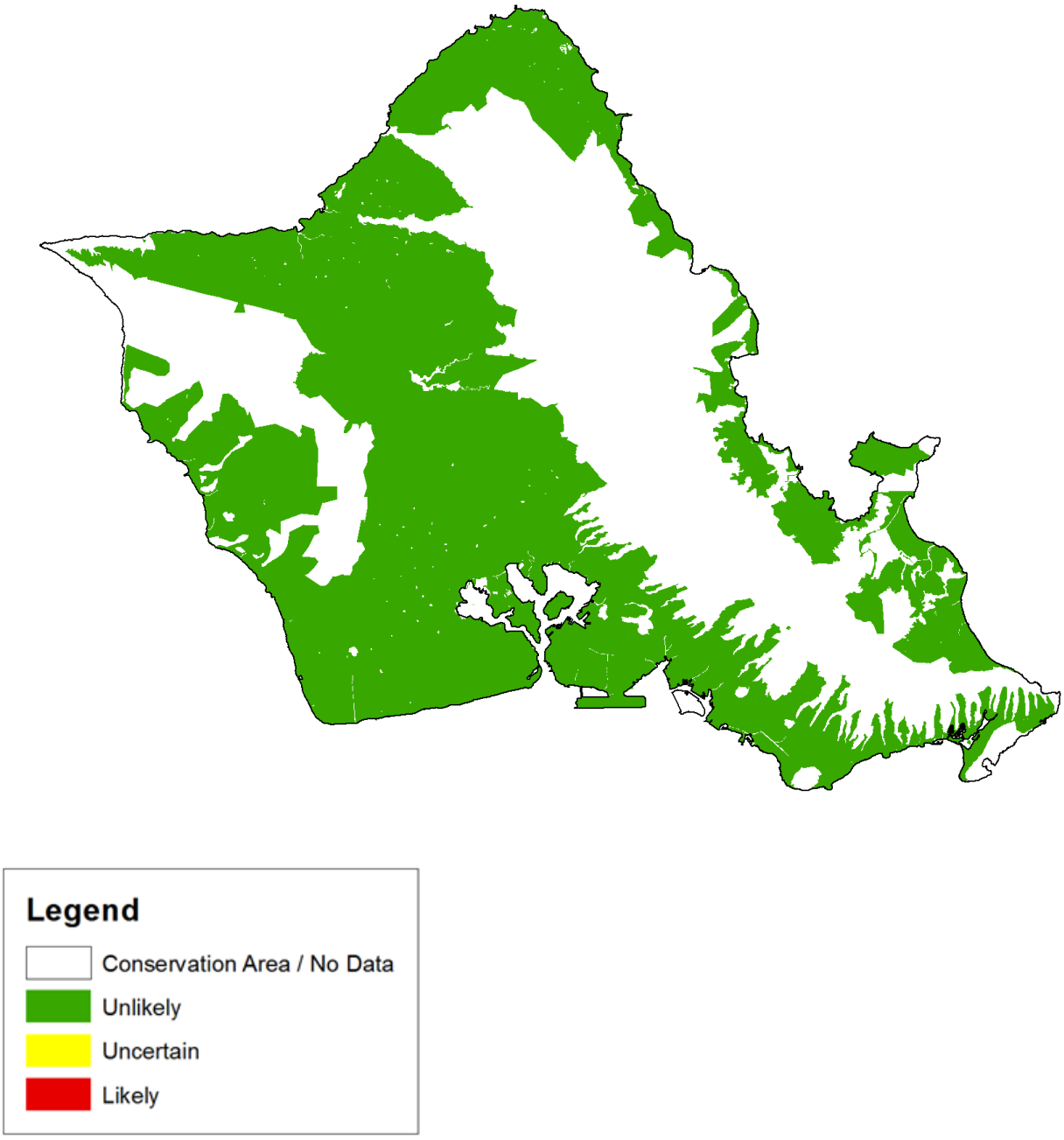
Carbamazepine



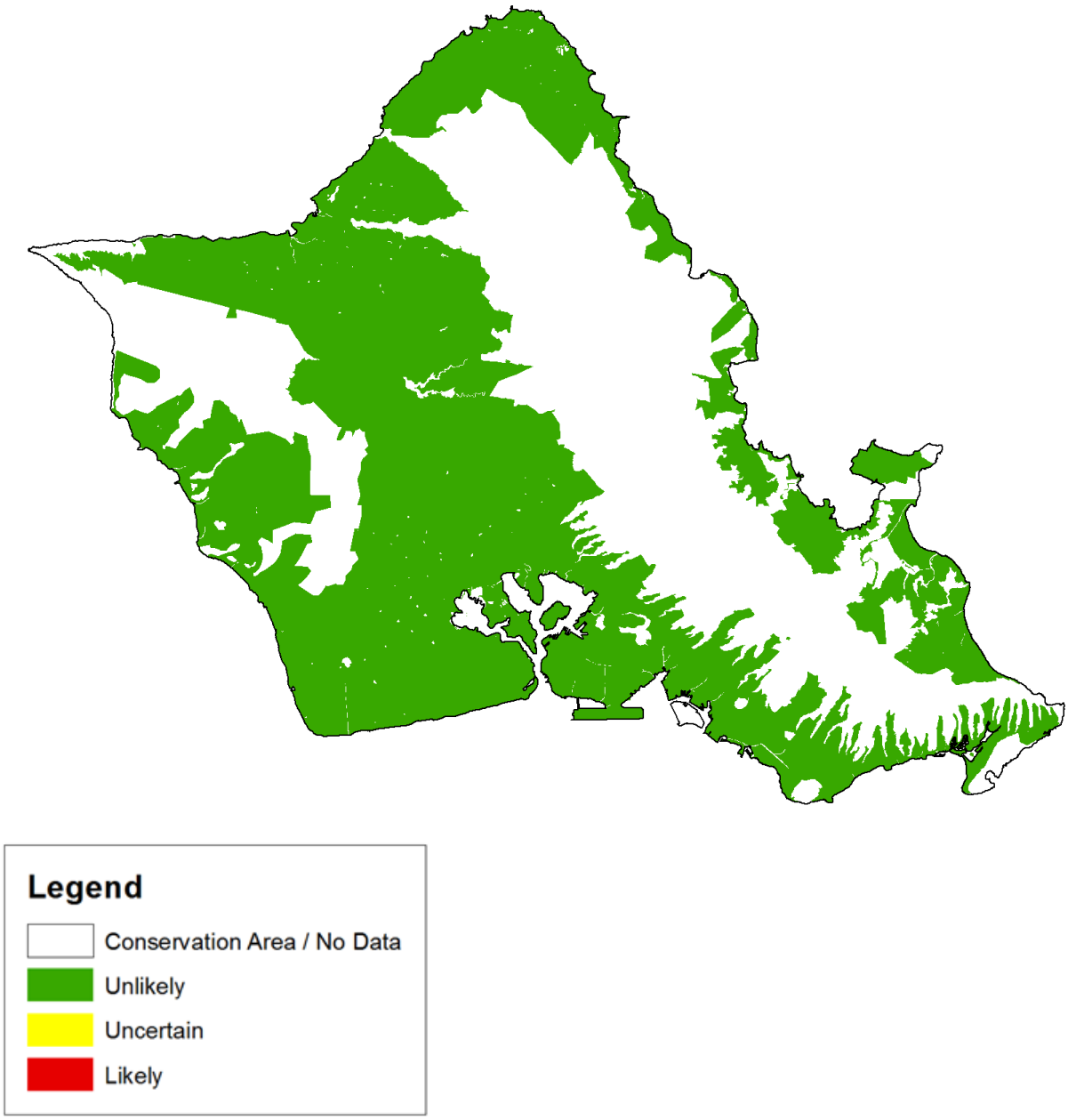
Estrone


Legend
Conservation Area / No Data
Unlikely
Uncertain
Likely

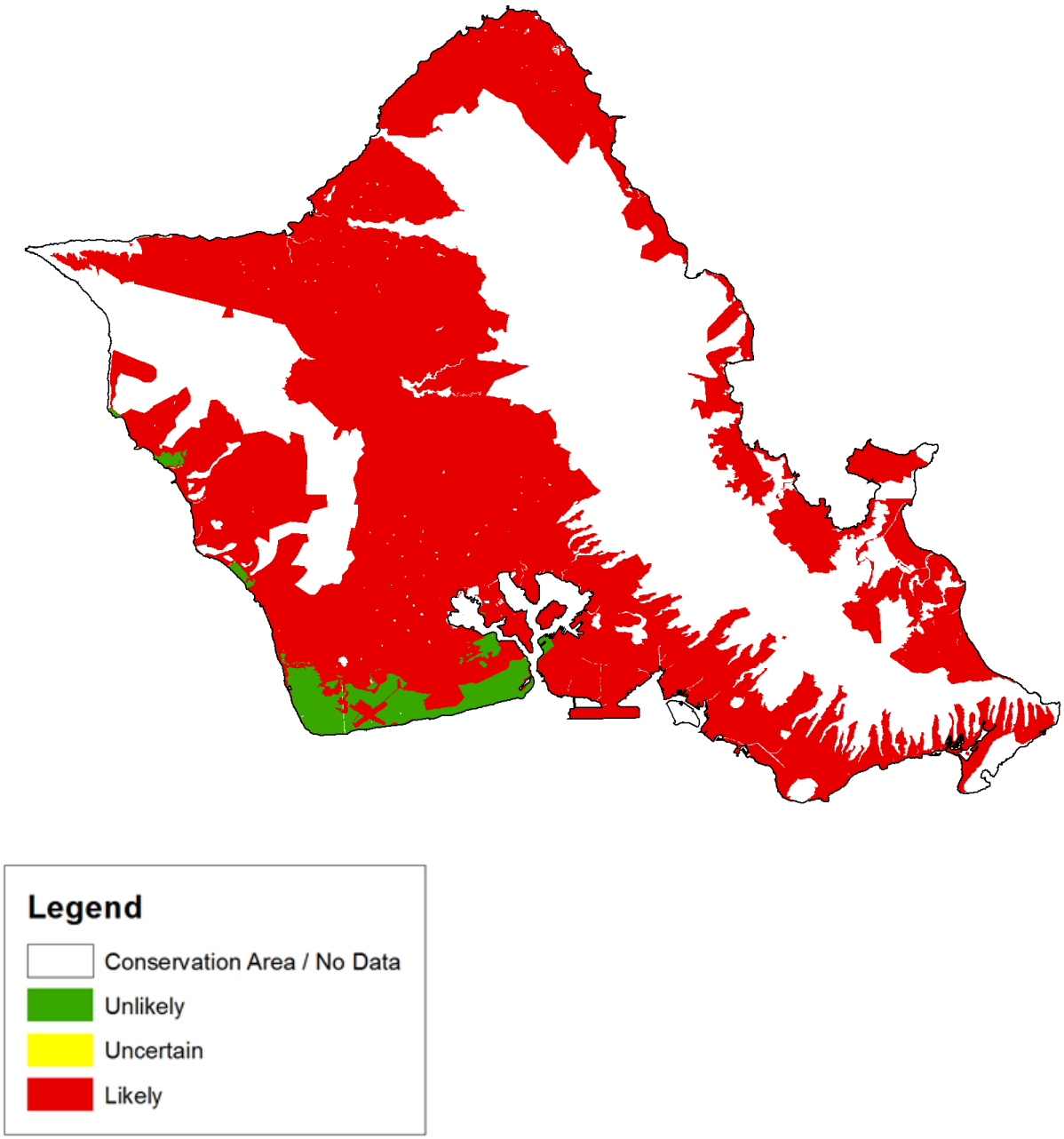
Estradiol


Legend
Conservation Area / No Data
Unlikely
Uncertain
Likely

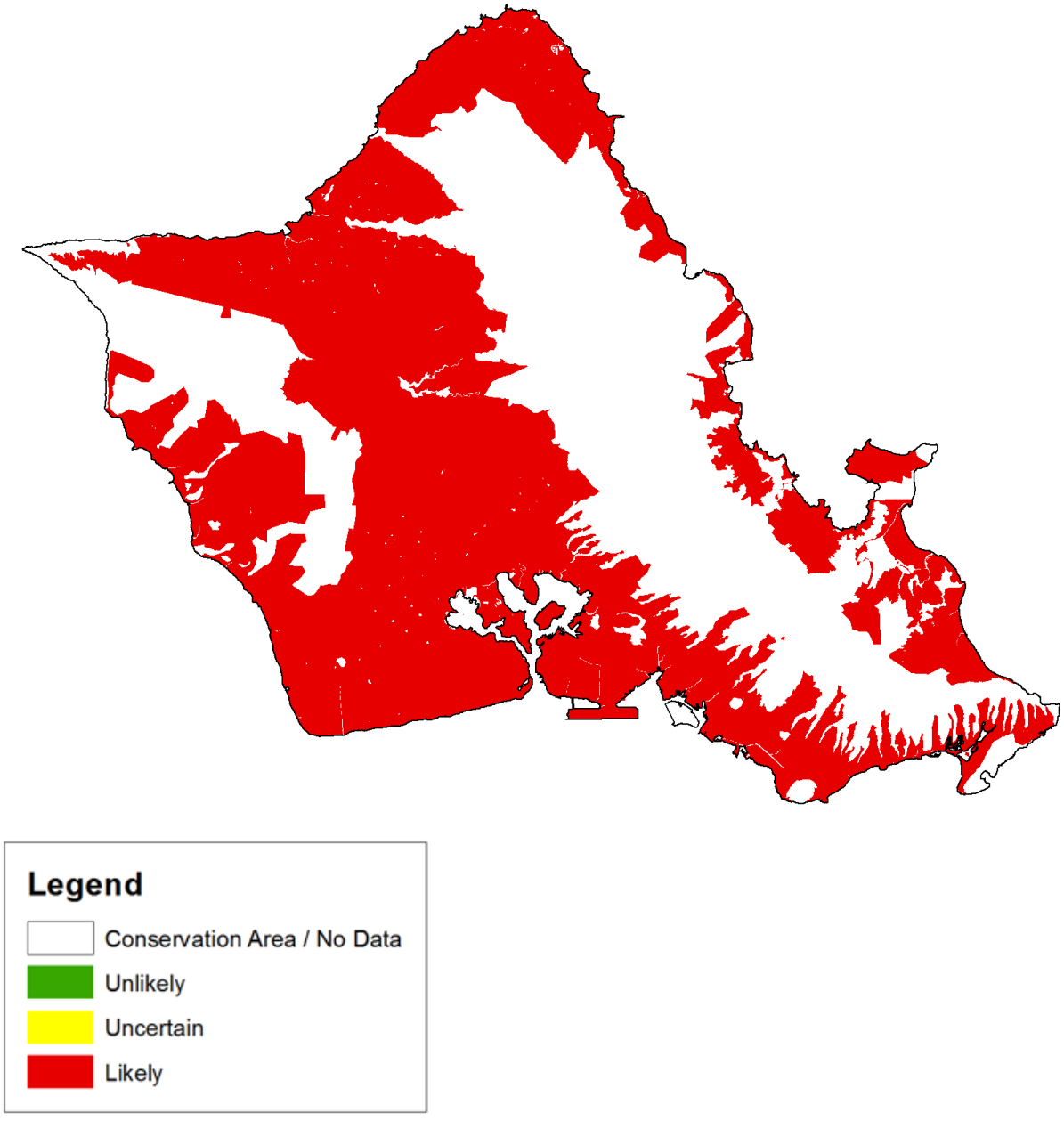
Ethinylestradiol



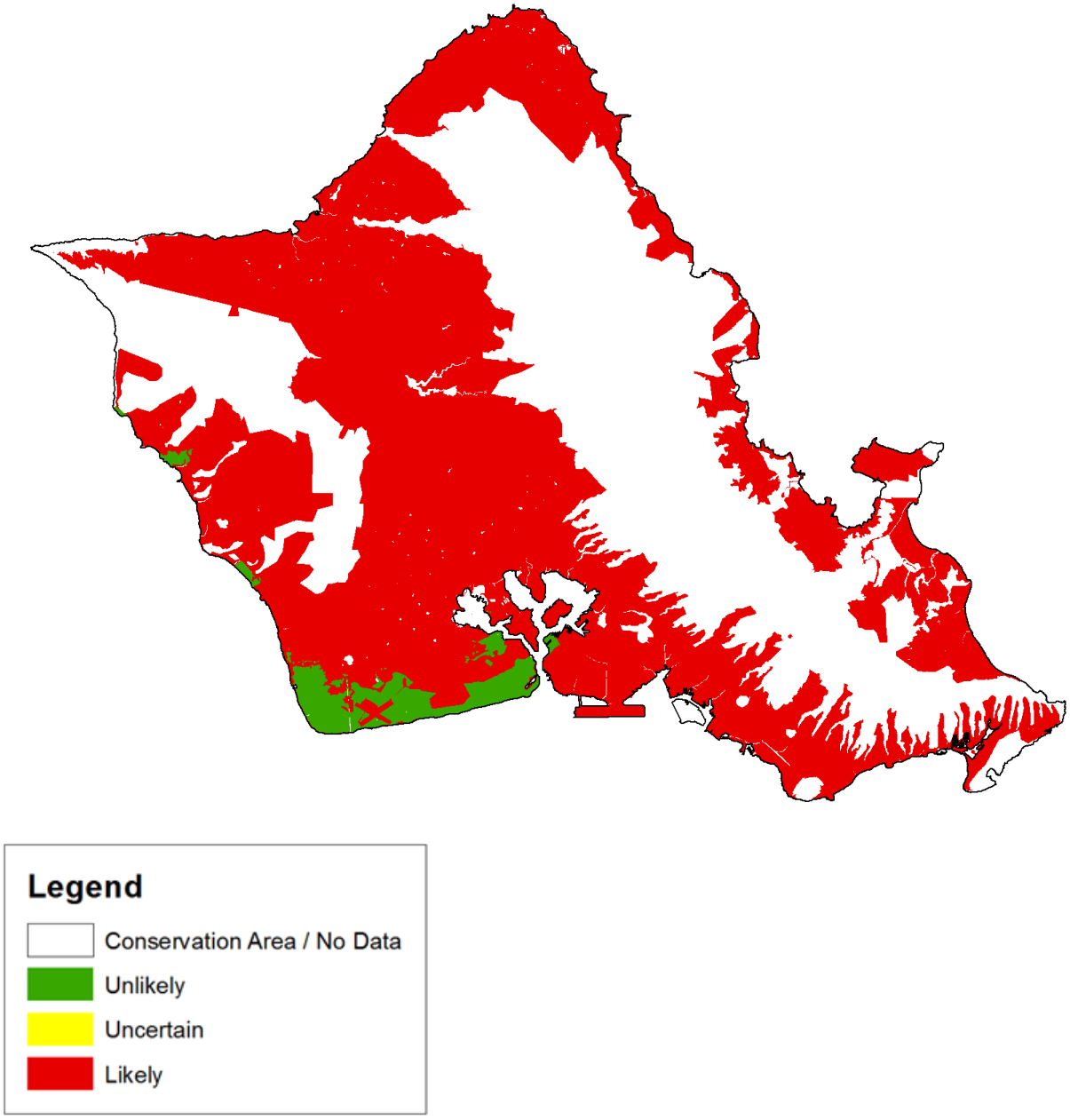
Azithromycin



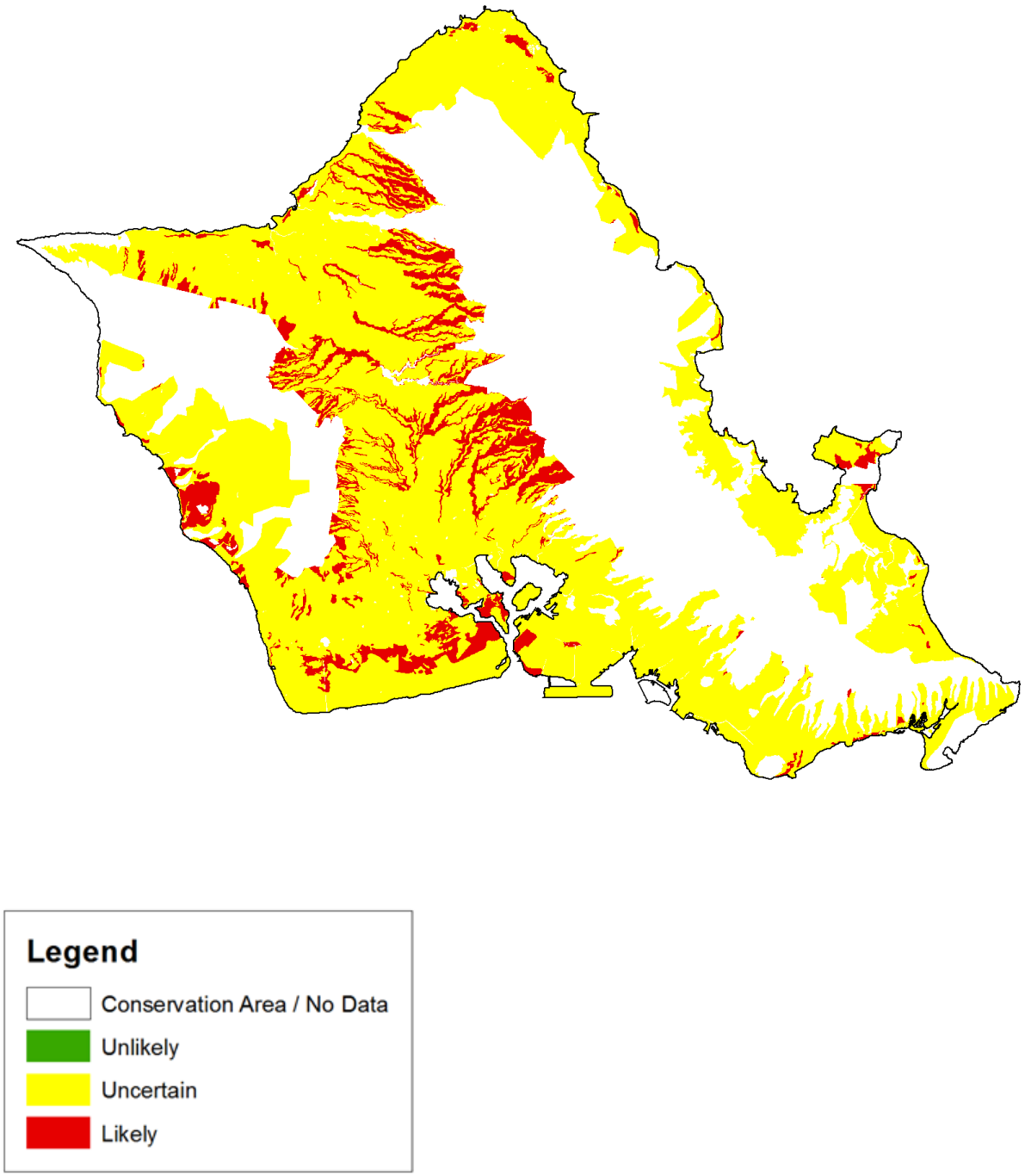
Clarithromycin



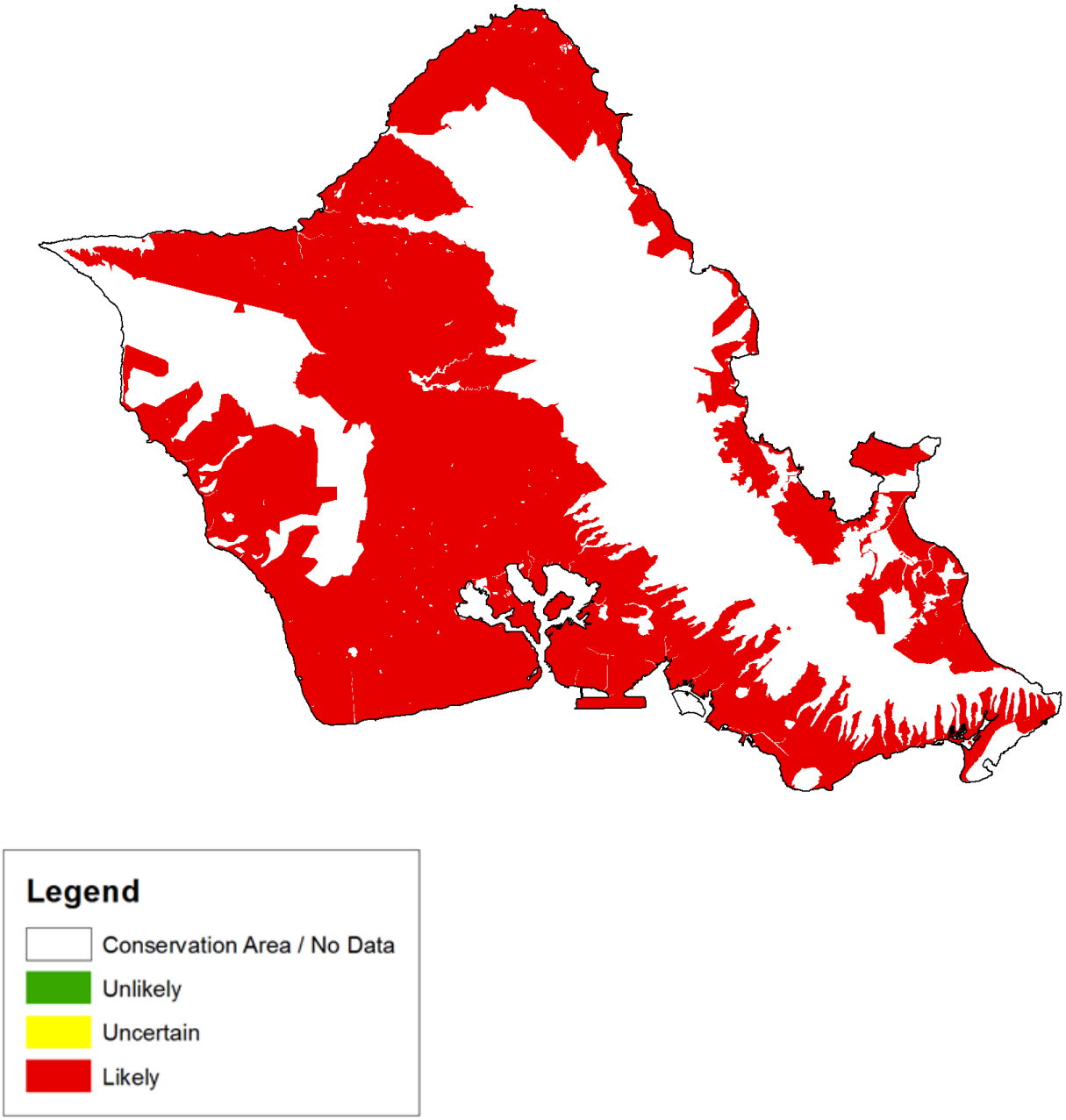
Roxithromycin



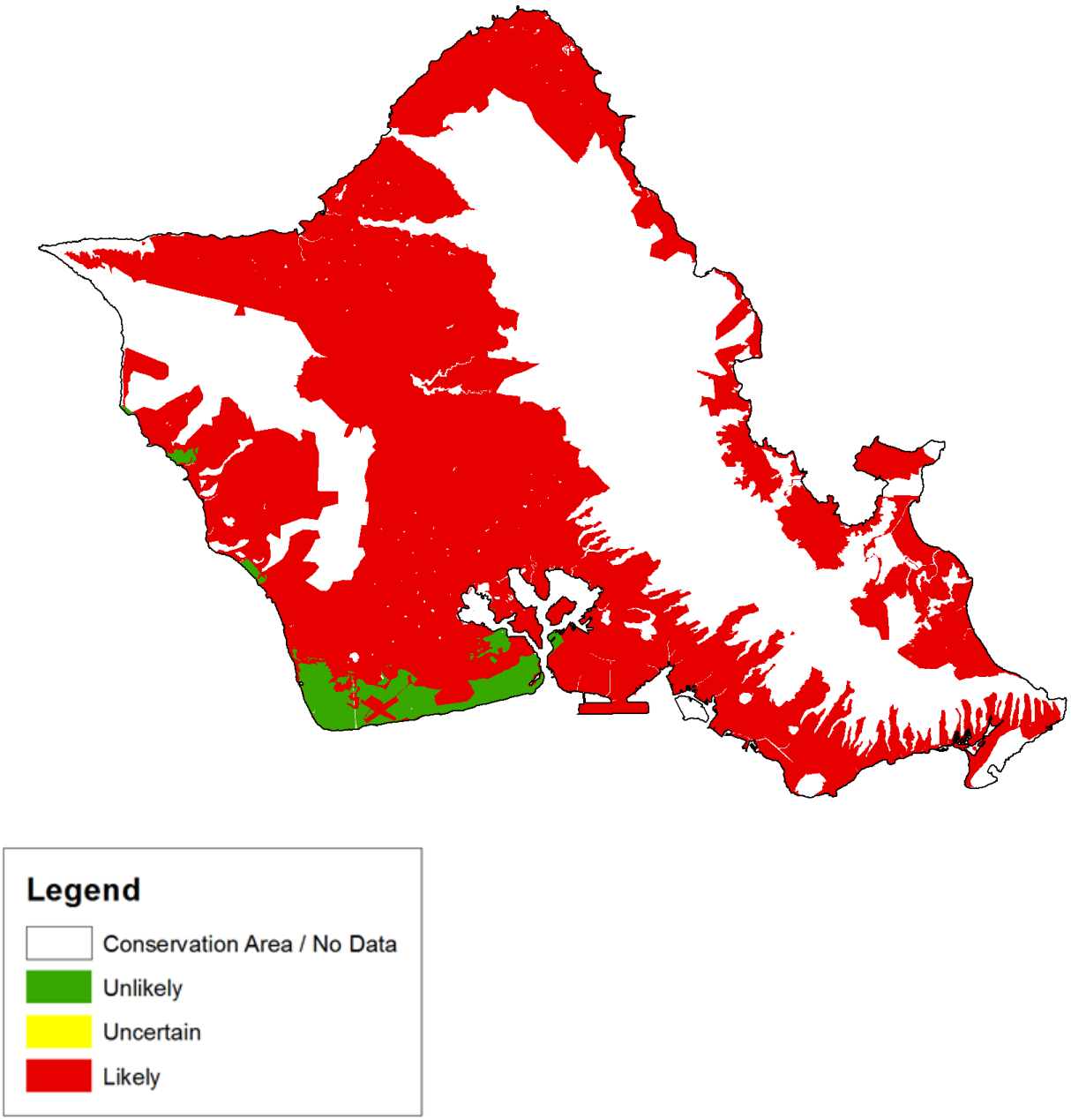
Sulfamethoxazole



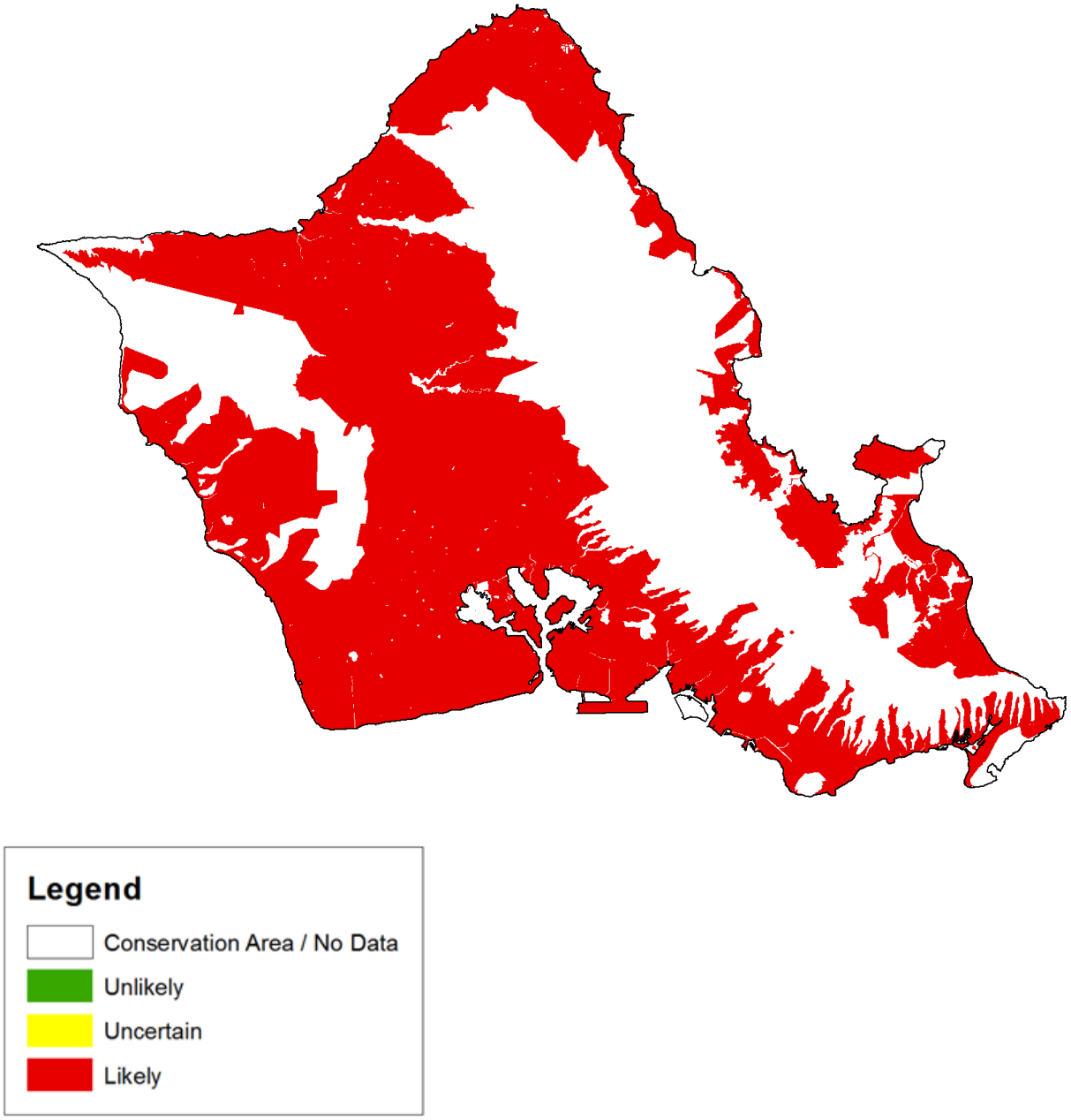
Ciprofloxacin



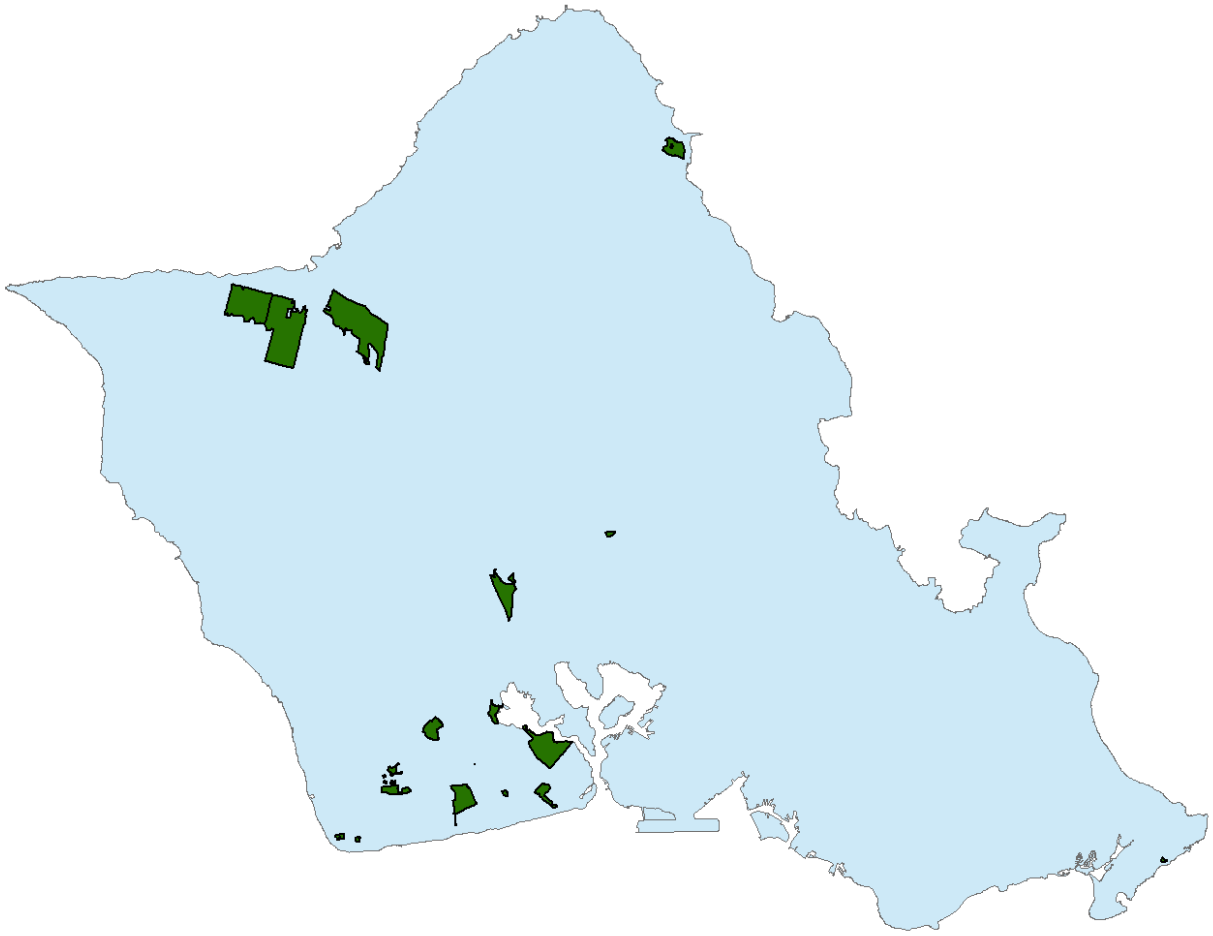
Ofloxacin


Legend
Conservation Area / No Data
Unlikely
Uncertain
Likely

Propranolol



Appendix C – Tax Map Keys that reuse wastewater on Oahu



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