ÉTUDE DES IMPACTS ANTHROPIQUES DANS LES LACS CANADIENS : ANALYSE DES CONTAMINANTS ORGANIQUES À L'ÉTAT DE TRACE

par

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SOMMAIRE

Les lacs sont des écosystèmes complexes apportant de nombreux avantages tant pour l'environnement que pour l'être humain. Au Canada, les lacs ont une importance d'autant plus grande qu'ils sont plus de 2,4 millions présents à travers son territoire. Cependant, ces écosystèmes sont sensibles aux activités anthropiques prenant place dans leurs bassins versants et ils peuvent être des puits de contamination du fait des rejets humains. En effet, de nombreux composés chimiques sont produits et utilisés quotidiennement, et ces contaminants se répandent souvent dans les eaux de surface naturelles. Toutefois, il y a un manque de données homogènes provenant des méthodologies comparables sur la répartition de la contamination anthropique à grande échelle dans les lacs. Il est donc nécessaire de combler cette lacune tout en étudiant l'impact de cette contamination et ses sources. L'objectif général du projet présenté ici est d'évaluer la présence de certains de ces contaminants dans les lacs canadiens, leurs potentiels impacts sur le biote lacustre ainsi que les facteurs menant à cette contamination.

Cette thèse de doctorat porte donc sur l'analyse de la présence de contaminants organiques à l'état de traces (TrOCs) dans les lacs canadiens. Pour cela, une méthode d'analyse ciblée permettant la quantification dans l'eau de lac de 54 TrOCs, représentatifs de diverses activités humaines, a été développée. Les échantillons sont d'abord concentrés par extraction sur phase solide avant analyse par chromatographie liquide couplée à la spectrométrie de masse en tandem.

En premier lieu, la présence des contaminants a été étudiée dans les eaux de surface de 290 lacs à travers le Canada (Chapitre 2). Des contaminants ont été détectés dans 88 % des lacs échantillonnés, avec jusqu'à 28 détections par lac. Les TrOCs ont été quantifiés jusqu'à environ 2 200 ng/L pour les composés individuels, avec des concentrations cumulatives allant jusqu'à plus de 8 100 ng/L dans l'eau de lac. Une évaluation du risque environnemental des TrOCs retrouvés a été réalisée pour trois espèces aquatiques (*Pimephales promelas*, *Daphnia magna* et *Tetrahymena pyriformis*) en comparant les concentrations environnementales aux seuils de toxicité des différents contaminants pour ces espèces modèles représentant différents niveaux trophiques. Les résultats ont révélé que 6 % des lacs présentaient un risque élevé pour au moins une de ces espèces. Dans 59 % des lacs, certains contaminants ayant des effets sous-létaux potentiels ont été détectés.

Ces TrOCs peuvent avoir diverses sources dépendant principalement du type d'activités humaines prenant place dans son bassin versant. Des études exploratoires ont mis en évidence des facteurs potentiels de cette contamination. Le deuxième objectif de cette étude était donc de vérifier les effets de ces facteurs sur la contamination de l'eau (Chapitre 3). Pour ce faire, nous avons relié les données de détection et de concentrations des TrOCs à des données d'activités et d'utilisations du sol d'origine anthropique à l'aide de modèles de régression linéaire et logistique. Ces modèles ont été choisis afin de permettre la prise en compte de la grande proportion de concentrations inférieures aux limites de la méthode. Malgré l'effet potentiel de nombreux paramètres sur l'atténuation de la contamination, les analyses ont confirmé l'impact prédominant de l'utilisation des terres agricoles et urbaines et des principales sources ponctuelles anthropiques de contaminants, tels que la présence de stations de traitement des eaux usées, dans les bassins versants sur la contamination organique trace des lacs. Ces résultats peuvent permettre d'améliorer les prises de décisions concernant la réduction des sources de contamination des lacs en réduisant le nombre de variables à considérer. Des recommandations ont également été émise pour le suivi de lacs ayant de plus grands risques de contamination à travers la Canada.

Ces travaux constituent le premier point de référence pour le suivi de l'évolution de la contamination des lacs canadiens par les TrOCs. Ils démontrent qu'une grande partie des lacs échantillonnés portent une empreinte chimique d'origine humaine pouvant entraîner des conséquences importantes pour l'environnement, et l'ensemble de ces résultats peut conduire à une meilleure gestion des bassins versants. Par la suite, ces résultats pourront être mis en relation avec d'autres données ayant été acquises pour ces lacs par d'autres groupes de recherche, telles que la présence de gènes de résistance aux antibiotiques ou la structure des communautés planctoniques, afin de mieux comprendre les impacts des TrOCs sur le fonctionnement des écosystèmes lacustres.

Mots clés : Écosystèmes aquatiques, Lacs, Impacts anthropiques, Contaminants organiques à l'état de traces, Pharmaceutiques, Pesticides, Écotoxicité, Facteurs de contamination.

ABSTRACT

Lakes are complex ecosystems that provide countless benefits for both the environment and humans. In Canada, lakes are especially important as there are over 2.4 million of them throughout the country. However, these ecosystems are sensitive to the human activities taking place in their watersheds, and they can be sinks of contamination led by human discharges. Indeed, many chemical compounds are produced and used daily, and these contaminants often find their way into natural surface waters. However, there is a lack of homogeneous data from comparable methodologies on the distribution of large-scale anthropogenic contamination in lakes. Therefore, it is necessary to fill this gap by studying the impacts and sources of this contamination. The general objective of the project presented here is to assess the presence of some of these contaminants in Canadian lakes, their potential impact on lake biota and the factors leading to this contamination.

This doctoral thesis thus focuses on the analysis of the presence of trace organic contaminants (TrOCs) in Canadian lakes. To this end, a targeted analytical method for quantifying 54 TrOCs, representative of various human activities, in lake water was developed. Samples are first concentrated by solid-phase extraction before analysis by liquid chromatography coupled with tandem mass spectrometry.

Firstly, the presence of contaminants was studied in the surface waters of 290 lakes across Canada (Chapter 2). Contaminants were detected in 88% of the lakes sampled, with up to 28 detections per lake. TrOCs were quantified up to around 2,200 ng/L for individual compounds, with cumulative concentrations up to over 8,100 ng/L in sampled lake water. An environmental risk assessment of these TrOCs was carried out for three aquatic species (Pimephales promelas, Daphnia magna and Tetrahymena pyriformis) by comparing environmental concentrations with the toxicity thresholds of the various contaminants for these species. The results revealed that 6% of lakes presented a high risk for at least one of these species. In 59% of lakes, some contaminants with potential sub-lethal effects were detected.

These TrOCs can come from a variety of sources, depending mainly on the type of human activity taking place in their watersheds. Exploratory studies have highlighted potential factors of this contamination. The second objective of this study was thus to verify the effects of these factors on water contamination (Chapter 3). To do so, we linked TrOCs detection and concentration data to anthropogenic land use and activity data using linear and logistic regression models. These models were chosen to take into account the large proportion of concentrations below the limits of the method. Despite the potential effect of numerous parameters on contamination attenuation, the analyses confirmed the predominant impact of agricultural and urban land use and major anthropogenic point sources of contaminants, such as the presence of wastewater treatment plants, in watersheds on trace organic contamination of lakes. These results can help improve decision-making regarding the reduction of lake contamination sources by decreasing the number of variables to consider. Recommendations were also given to monitor lakes with higher risks of contamination across Canada.

This work represents the first reference point for monitoring the evolution of TrOCs contamination in Canadian lakes. They demonstrate that a large proportion of the sampled lakes display an anthropogenic chemical footprint that can lead to significant consequences for the environment, and together these results can lead to better watershed management. These results could then be linked to other data acquired for these lakes by other research groups, such as the presence of antibiotic resistance genes or the structure of planktonic communities, to better understand the impact of TrOCs on the functioning of lake ecosystems.

Key words: Aquatic ecosystems, Lakes, Anthropogenic impacts, Trace organic contaminants, Pharmaceuticals, Pesticides, Ecotoxicity, Contamination factors.

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2,4-D	Acide 2,4-dichlorophénoxyacétique
95%CI	Intervalle de confiance de 95%
BPA	Bisphénol A
CE_{50}, EC_{50}	Concentration effective médiane
CIE	Contaminants d'intérêt émergent
CL_{50}, LC_{50}	Concentration létale médiane
DEET	Diéthyltoluamide ou N,N-Diéthyl-3-méthylbenzamide
IGC_{50}	Concentration médiane d'inhibition de la croissance
IRR	Rapport de taux d'incidence
LC-MS/MS	Chromatographie liquide couplée à la spectrométrie de masse en tandem
LOD	Limite de détection
LOQ	Limite de quantification
MCPA	Acide 2-méthyl-4-chlorophénoxyacétique
OR	Rapport de cote
PNEC	Concentration sans effet prédite
POP	Polluants organiques persistants
PPCPs	Produits pharmaceutiques et de soin personnel
QR, RQ	Quotient de risque
QSAR	*Quantitative Structure–Activity Relationship*
SPE	Extraction en phase solide
STEP, WWTP	Station d'épuration des eaux usées
T.E.S.T.	*Toxicity Estimation Software Tool*
TBEP	Phosphate de tris(2-butoxyéthyle)
TBP	Phosphate de tributyle
TCPP	Tris(1-chloro-2-propyle)
TDCPP	Phosphate de tris(dichloroisopropyle)
TPP	Phosphate de triphényle
TrOCs	Contaminants organiques à l'état de trace

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CHAPITRE 1. INTRODUCTION

Depuis des décennies, voire des siècles, les pressions humaines sur l'environnement ne cessent de croître. L'être humain, en cherchant à améliorer sa condition et son confort de vie, utilise la Nature et la souille sans réaliser les retombées de ses actes. Les conséquences de ses actions se font sentir dans chaque compartiment environnemental, de l'atmosphère à la terre, en passant par les écosystèmes aquatiques (Steffen et al., 2007). Cependant, l'eau est une de nos ressources les plus précieuses et les plus fragiles. Nos impacts sur ce compartiment environnemental entraînent des conséquences importantes qui continueront d'augmenter dans le futur par l'augmentation des pressions anthropiques et des dérèglements, notamment climatiques, qui en découlent. Il est donc très important de comprendre les impacts humains sur les ressources en eau pour pouvoir limiter leurs conséquences et orienter les efforts de conservation de ces milieux. Dans cette thèse, nous nous intéresserons aux écosystèmes aquatiques complexes que sont les lacs. En effet, malgré leur importance, la contamination des lacs est peu étudiée en comparaison des rivières (Petrie et al., 2015). Or, les lacs sont sensibles à la contamination liée aux pressions anthropiques, notamment par des composés organiques qui peuvent se montrer toxiques pour les organismes aquatiques. Il est donc nécessaire de porter notre attention sur ces ressources afin de mieux les protéger.

1.1. Lacs

1.1.1. Caractéristiques des lacs

Un lac est globalement défini comme un plan d'eau continental n'étant pas relié directement à une mer ou un océan (Touchart, 2000). Les lacs sont des plans d'eau stagnante. Ils sont donc caractérisés par un flux très lent, ce qui entraîne un temps de résidence plus long que pour les eaux courantes telles que les rivières. La stagnation de ces eaux mène également au développement d'une stratification thermique lorsque la profondeur du lac est suffisante. Cette stratification est composée d'une couche supérieure, l'épilimnion, et d'une couche inférieure, l'hypolimnion, qui sont séparées par une couche de transition appelée métalimnion. L'épilimnion et l'hypolimnion sont des couches mélangées et plutôt homogènes, tandis que le métalimnion contient un gradient de température. Cette stratification est en place en été, avec un épilimnion plus chaud et un hypolimnion froid, et en hiver, dans les pays de grands froids comme le Canada, avec un épilimnion à des températures proches de 0 °C et un hypolimnion aux alentours de 4 °C. Dans ce cas, les lacs subissent un mélange tout au long de leur colonne d'eau à l'automne et au printemps, pendant lequel il n'y a plus de stratification marquée.

Les lacs sont des systèmes très variés, en premier lieu par leurs dimensions. Ils peuvent atteindre des surfaces équivalentes à des mers intérieures de plusieurs milliers de kilomètres carrés (km^2) ; cependant, les lacs les plus abondants ont une surface inférieure à 1 km^2 (Messager et al., 2016). La limite de taille inférieure permettant de considérer une étendue d'eau comme un lac n'est pas claire, mais elle sera fixée dans ce travail à 0,1 km^2, une valeur seuil souvent utilisée dans la littérature. Leur profondeur varie également, pouvant atteindre plusieurs centaines de mètres dans certaines régions. En effet, la profondeur d'un lac est reliée à la géographie environnante et les lacs de zones montagneuses sont souvent plus profonds que les lacs de plaines.

Une caractéristique importante des lacs est l'incorporation de tout ce qui se déroule dans leurs bassins d'alimentation, ou bassins versants. Le bassin versant d'un plan d'eau est la zone géographique dans laquelle chaque goutte d'eau qui tombe finit par atteindre ce système aquatique (**Figure 1**). En tant que réservoirs où converge toute l'eau d'un bassin versant, les lacs intègrent donc la contamination provenant de sources diffuses (e.g., le ruissellement) et ponctuelles (e.g., les effluents de stations d'épuration des eaux usées (STEP)). Les lacs sont fortement influencés par leurs bassins versants, et cela est d'autant plus marqué que le ratio entre l'aire du bassin et l'aire du lac est élevé (Touchart, 2000).

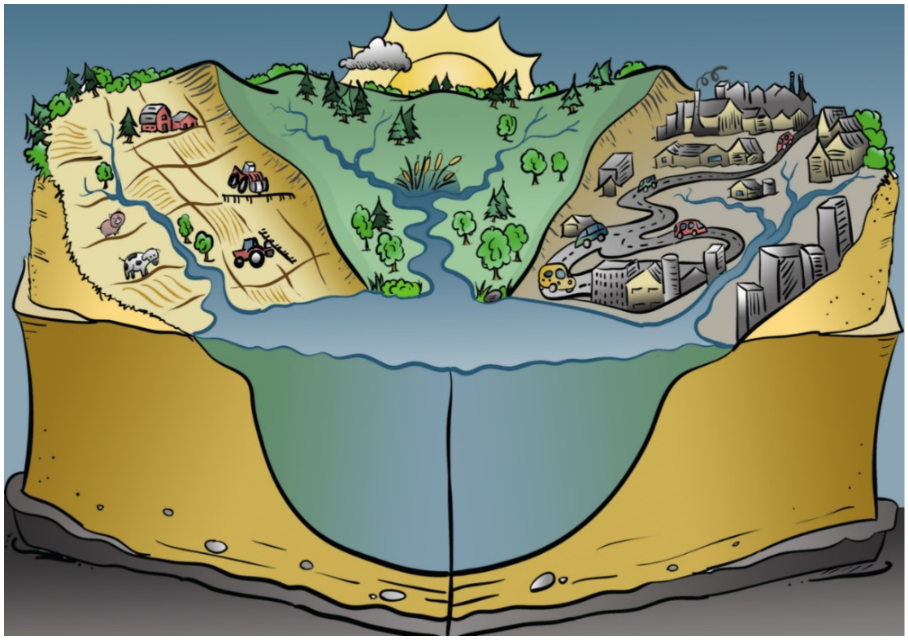

Figure 1. Représentation d'un bassin versant. Reproduite avec la permission du réseau Lake Pulse. Copyright 2024 Catherine A. Brown.

1.1.2. Importance des lacs

Les lacs couvrent environ 1,8% de la surface continentale mondiale (Lehner and Döll, 2004; Messager et al., 2016) et contiennent 87% de l'eau douce de surface non gelée (Yao et al., 2023). Les lacs sont donc une importante ressource en eau douce à travers le monde. Il est possible de déterminer l'importance de ces écosystèmes en utilisant la notion de services écosystémiques, définis comme les bénéfices et services essentiels prodigués par les écosystèmes (Inácio et al., 2022; Reynaud and Lanzanova, 2017). Ce concept permet d'évaluer l'importance des écosystèmes à l'échelle locale ainsi que de quantifier les potentiels impacts de leur dégradation. Ces services entrent dans deux grandes catégories, les services pour l'humain et les services pour l'environnement. Les principaux services rendus aux humains par les lacs sont : une importante source pour l'eau potable et l'irrigation, le support de la pisciculture, la production d'électricité, et le lieu de nombreuses activités récréatives, culturelles et spirituelles. Les principaux services liés à l'environnement sont : la régulation des cycles biogéochimiques (en particulier ceux du carbone, du phosphore et de l'azote), la régulation du climat à l'échelle régionale, la limitation des inondations extrêmes, et le maintien de la biodiversité. Les lacs sont donc des ressources inestimables, toutefois, encore trop peu d'études portent sur l'analyse de la qualité de ces systèmes complexes (Konstantinou et al., 2006; Petrie et al., 2015).

L'importance des lacs dans le maintien de la biodiversité s'applique non seulement aux organismes aquatiques mais aussi aux organismes terrestres. En effet, ces écosystèmes sont des habitats privilégiés pour les espèces lacustres y vivant en partie, tels que les amphibiens, les insectes et certains reptiles et oiseaux, ou y demeurant entièrement, tels que les poissons, les plantes aquatiques, le zooplancton (e.g., crustacés, mollusques), et le phytoplancton (e.g., cyanobactéries, algues, diatomées). Cependant, ils peuvent également être une source de nourriture importante pour des espèces terrestres tels que des oiseaux et mammifères piscivores (Adrian et al., 2016; Dudgeon et al., 2006).

Les lacs sont une ressource particulièrement importante au Canada qui compterait plus de 2,4 millions de lacs sur son territoire (Cooke and Murchie, 2015), dont près d'1 million sont de taille supérieure à 0,1 km^2 (Messager et al., 2016). C'est le pays comportant le plus de lacs en nombre (Sayers et al., 2015) (**Figure 2**) mais également en surface puisque les lacs canadiens représentent 37% de la surface lacustre mondiale (Minns et al., 2008). Cependant, les efforts de recherche sur les lacs au Canada portent principalement sur les lacs de très grande taille comme les Grands Lacs, le Lac Winnipeg ou le Grand Lac des Esclaves. Les lacs de plus petites tailles, pourtant plus nombreux et distribués plus largement sur le territoire, reçoivent, eux, moins d'attention. Ces lacs ont néanmoins une grande importance locale, notamment pour les peuples indigènes.

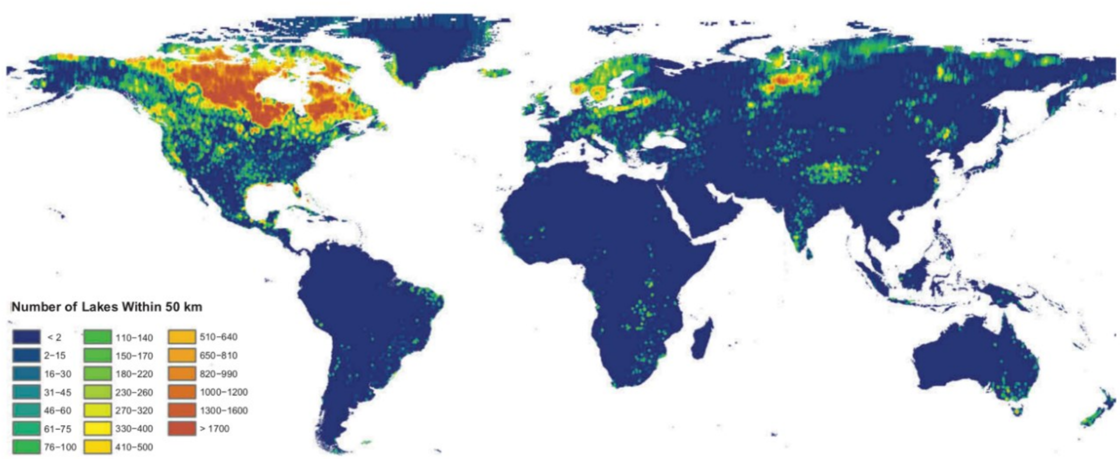

Figure 2. Distribution du nombre de lacs dans le monde. Reproduit avec la permission de Informa UK Limited, trading as Taylor & Taylor & Francis Group, http://www.tandfonline.com. Sayers et al., 2015. A new method to generate a high-resolution global distribution map of lake chlorophyll. *International Journal of Remote Sensing, 36*(7), 1942–1964. Copyright tres/03863768 [2024].

C'est dans ce contexte qu'a pris forme le réseau du Conseil de recherches en sciences naturelles et en génie du Canada (CRSNG) sur l'état des lacs du Canada (appelé « Lake Pulse » dans la suite de ce document), dans lequel s'inscrit une partie de ma recherche. Les objectifs du projet Lake Pulse sont de combler le manque d'information sur l'état de santé des lacs canadiens, de développer de nouveaux indicateurs de fonctionnement des écosystèmes et d'élaborer des scénarios de changement dans les écosystèmes aquatiques (Huot et al., 2019).

1.2. Contamination environnementale

Les pressions anthropiques exercées sur l'environnement peuvent mener à la dégradation des écosystèmes par l'apparition de contamination tant biologique que chimique. La contamination peut être décrite comme la présence ou l'introduction d'un élément en quantité supérieure à celle attendue naturellement dans l'environnement (Chapman, 2007). Les principales contaminations biologiques sont : les espèces envahissantes, se propageant de lac en lac par des embarcations récréatives mal nettoyées (Vander Zanden and Olden, 2008) et pouvant mener à un déséquilibre de la biodiversité lacustre (Havel et al., 2015; Walsh et al., 2016) ; les pathogènes, provenant du bassin versant ou de l'utilisation directe du lacs par des oiseaux et autres animaux (Brookes et al., 2004; Pandey et al., 2014) et risquant d'impacter la biodiversité de ces écosystèmes et leur potabilité ; les efflorescences algales, qui produisent des toxines et dont la prolifération mène à une forte diminution de la saturation en oxygène, létale pour certains poissons et autres espèces aquatiques (Huisman et al., 2018). Ces dernières peuvent être dues à un apport important de nutriments favorisant la prolifération d'algues. Ce phénomène s'appelle l'eutrophisation et il est principalement lié à l'épandage d'azote et de phosphore sur les terres agricoles (Kakade et al., 2021). Cet excès de nutriments est une première contamination chimique possible.

Les autres contaminations chimiques pouvant impacter les lacs sont notamment : les métaux d'origine naturelle ou anthropique, en particulier les éléments-traces métalliques tels que le cadmium, le nickel, le plomb ou le mercure, qui proviennent du développement industriel et minier ou de leur application comme fertilisants ou pesticides, et qui posent un risque pour les organismes aquatiques (Karaouzas et al., 2021) ; les sels de voirie tels que le chlorure de sodium, le chlorure de magnésium et le chlorure de calcium, abondamment utilisés au Canada pour le déglaçage des routes en hiver (Government of Canada, 2018) et dont la présence dans les lacs impactent autant la stratification (Dupuis et al., 2019) que les organismes aquatiques (Arnott et al., 2020; Hintz and Relyea, 2019) ; les microplastiques, regroupant les débris plastiques de taille inférieure à 5 mm (Arthur et al., 2009) utilisés directement sous cette forme ou provenant de la dégradation de plus gros plastiques (D'Avignon et al., 2022), qui peuvent bloquer le système digestif des organismes aquatiques ou libérer des composés adsorbés (Bejgarn et al., 2015; Setälä et al., 2016) ; les composés organiques, regroupant différentes catégories, qui sont l'objet de cette recherche.

1.2.1. Contaminants organiques à l'état de trace

Les premiers composés organiques dont la présence environnementale à fait l'objet de recherches sont les polluants organiques persistants (POP). Ces composés sont particulièrement problématiques car ils se répandent dans différents compartiments environnementaux et peuvent y rester intacts durant de longues périodes (Xu et al., 2013). Ils sont alors souvent bioaccumulés et exercent une grande toxicité tant pour la faune aquatique que pour l'homme. Cette catégorie comprend notamment des composés industriels (polychlorobiphényles ou PCB), des retardateurs de flammes (polybromo-diphényléthers ou PBDE), des pesticides organochlorés (dichlorodiphényltrichloroéthane ou DDT, chlordanes, lindane) et des hydrocarbures aromatiques polycycliques (HAP) ; plus récemment ont été ajoutés des composés perfluorés (Houde et al., 2008; Li et al., 2019).

Cependant, la prise de conscience des risques pour l'Homme et l'environnement liés aux POP a conduit à la mise en place de réglementation pour ces composés, et des réglementations internationales ont été établies. En particulier, le protocole de Montréal relatif aux substances qui appauvrissent la couche d'ozone (SACO) de 1987 prévoyait l'élimination des SACO, dont les chlorofluorocarbures. La convention de Stockholm sur les polluants organiques persistants de 2001 a quant à elle donné lieu à un bannissement de substances dangereuses tels l'insecticide DDT. Des recommandations locales ont également vu le jour. En Europe, une directive pour l'eau à destination de la consommation humaine (Council of the European Union, 1998) ainsi qu'une directive-cadre sur l'eau visant la protection des eaux de surface (European Parliament and Council of the European Union, 2000) ont été produites. Aux États-Unis, une loi sur la protection de l'eau (Clean Water Act) a été mise en place, permettant à l'Agence de protection de l'environnement (US EPA, de l'anglais *United States Environmental Protection Agency*) d'établir une liste des critères de qualité de l'eau, notamment pour la vie aquatique (US EPA, 2015). Au Canada, il n'existe pas de réglementation sur la qualité des eaux de surface mais des recommandations ont été établies par le Conseil canadien des ministres de l'environnement (CCME, 2007). Cependant, peu de contaminants sont inclus dans ces recommandations. Une liste de contaminants à surveiller dans les effluents de station d'épuration a néanmoins été établie par le Ministère de l'Environnement du Québec, mais celle-ci n'inclut pas de valeur seuil (MELCCFP, 2023). Des critères de qualité de l'eau de surface ont également été établis pour plus de 400 contaminants au Québec en se basant principalement sur les recommandations canadiennes (MELCC, 2021).

Les polluants réglementés par des conventions internationales ou des réglementations régionales font donc l'objet de suivis et leur production est souvent ralentie ou arrêtée. Cependant, de nouveaux composés sont synthétisés pour les remplacer. En effet, les substances chimiques sont développées dans le but d'améliorer le confort et de lutter contre des problèmes rencontrés dans nos sociétés. L'interdiction d'utiliser des composés ayant une utilité considérée comme importante par la société mène donc au développement et à la production de nouvelles molécules pouvant prendre leur place pour ces utilisations. Il y a aujourd'hui plus de 219 millions de substances enregistrées dans le registre du Chemical Abstracts Service (CAS, 2024), et le développement de nouveaux composés augmente de façon exponentielle (CAS, 2015). Par ailleurs, plus de 350 000 composés ou mélanges de composés ont été déclarés comme étant produit ou utilisé dans le monde (Wang et al., 2020). Or ces molécules peuvent être rejetées dans l'environnement, et finalement atteindre les eaux de surface, du fait de l'absence de réglementation sur leur production et leur utilisation.

C'est ainsi qu'est apparu le terme de contaminants d'intérêt émergent (CIE). Ce sont des composés naturels ou de synthèse dont la présence dans l'environnement a été récemment découverte, et qui présentent un danger pour les organismes vivants du fait de leur toxicité ou de leur persistance dans l'environnement (Sauvé and Desrosiers, 2014). Ces contaminants ne sont pas encore réglementés et ne sont donc pas soumis à des suivis ou des contrôles. La découverte de nouveaux CIE augmente sans cesse, car ils proviennent du développement de nouveaux composés, mais aussi de l'amélioration des méthodes d'analyse qui permettent de détecter des molécules à des concentrations de plus en plus faibles.

Les POP et les CIE peuvent être regroupés sous le terme de contaminants organiques à l'état de trace (TrOCs, de l'anglais *trace organic contaminants*). Cette catégorie inclus des composés tels que des résidus pharmaceutiques, des principes actifs de produits de soin personnel, des additifs alimentaires et industriels, ou encore des produits phytosanitaires (Fischer et al., 2017; Sousa et al., 2018). Ces substances sont très largement utilisées et rejetées soit directement dans l'environnement, soit par les rejets de stations de traitement d'eaux usées municipales et industrielles (Daughton, 2004; Pal et al., 2010; Yang et al., 2017). Ces contaminants sont ubiquitaires dans l'environnement, et ils sont habituellement retrouvés dans les eaux naturelles à de faibles concentrations, allant du ng/L au µg/L. Ils ont été observés dans les eaux de surface de tous les continents, allant jusqu'à la détection de TrOCs dans les eaux douces d'Antarctique (Cai et al., 2012; Esteban et al., 2016; Minella et al., 2016). En effet, malgré le développement limité de cette région isolée, des additifs industriels tels que des retardateurs de flamme et des tensioactifs ont été quantifiés à des concentrations allant jusqu'à 188 et 261 ng/L pour le phosphate de tris(chloroisopropyle) et le nonylphénol, respectivement. Cette contamination globale a été mise en évidence dans une étude des rivières à l'échelle mondiale (Wilkinson et al., 2022). Certains contaminants tels que la carbamazépine, la metformine et la caféine ont été détectés dans plus de la moitié des rivières étudiées. Bien que cette recherche ne porte pas sur l'étude des sédiments, il est important de noter que c'est un compartiment important dans la contamination. En effet, les sédiments peuvent stocker et relarguer des contaminants durant de longues périodes, participant ainsi à leur persistance dans l'environnement (Chiaia-Hernández et al., 2020). Des mélanges complexes de contaminants peuvent alors être rendus biodisponibles pour les organismes benthiques (Muz et al., 2020).

Peu d'études à grande échelle, couvrant un territoire aussi large que le Canada, portent sur les contaminants organiques dans les lacs. Certains groupes de recherche ont effectué des études à grande échelle dans des rivières aux États-Unis (Bradley et al., 2017; Focazio et al., 2008; Kolpin et al., 2002) et en Europe (Loos et al., 2009). Les TrOCs sont très suivis dans les eaux potables, les rivières et les eaux usées (Patel et al., 2019; Sta Ana et al., 2021), mais les données sont moins fréquentes concernant leur présence dans les lacs, bien qu'ils soient connus comme étant des puits pour ce qui est relâché dans leurs bassins versants (Müller et al., 1998). Les études portant sur la contamination des lacs se concentrent en général sur une classe spécifique de composés, comme les antibiotiques (Luo et al., 2023), les agents sucrants (Buerge et al., 2009; Fu et al., 2020; Perkola and Sainio, 2014) ou les herbicides (Müller et al., 1997; Peck et al., 2020). En effet, l'étude des lacs à la plus grande échelle à ce jour est le *National Lakes Assessment* (NLA) effectué par l'agence de protection de l'environnement des États-Unis sur plus de 1000 lacs. Cependant, ce suivi inclut l'analyse d'un TrOC seulement, l'herbicide atrazine (Peck et al., 2020). Les études comprenant une plus grande diversité de contaminants portent eux souvent sur un nombre limité de lacs. Ainsi, des articles qui incluent plusieurs classes de produits pharmaceutiques et de soin personnel (PPCPs, de l'anglais *pharmaceuticals and personal care products*) se sont intéressés à leur présence dans 4 lacs éloignés en Irlande (Aherne et al., 2023), dans un grand lac du nord de la Chine

(Zhang et al., 2018), dans 6 lacs urbains du centre de l'Inde (Archana et al., 2017), dans 3 lacs urbains au Vietnam (Tran et al., 2014) et dans un grand lac en Suède (Daneshvar et al., 2010). Des recherches incluant également d'autres TrOCs comme des pesticides et des additifs ont été menées dans d'autres pays sur 1 à 4 lacs par étude, en analysant entre 4 et 105 composés, en Suisse (Öllers et al., 2001), au Brésil (Sodré et al., 2018), en Arabie Saoudite (Picó et al., 2020), aux États-Unis (Sharma and Hanigan, 2021; Subedi et al., 2015) et en Suède (Malnes et al., 2022). Anagnostopoulpou et al. (2022) ont étudié une région plus étendue en Grèce qui incluait 18 lacs analysés pour plus de 470 TrOCs. La caféine, le diéthyltoluamide (DEET) et le retardateur de flammes phosphate de tris(1-chloro-2-propyle) (TCPP) ont été retrouvés dans tous les lacs étudiés à des concentrations allant au-delà d'1 µg/L pour le DEET. Une étude à plus grande échelle de TrOCs dans les lacs a été effectuée par Ferrey et al. (2015) qui ont analysé 125 composés dans 50 lacs du Minnesota, États-Unis. Ils y ont observé le plastifiant bisphénol A et le répulsif pour insectes DEET dans près de la moitié des lacs étudiés, à des concentrations allant jusqu'à 308 ng/L pour le DEET, et avec une concentration maximale retrouvée à 406 ng/L pour l'hormone androstérone.

Cependant, au Canada, il y a un manque de données pour les TrOCs dans les eaux de surface (Anderson et al., 2015). Il existe tout de même des études sur ces contaminants, mais celles-ci se concentrent sur une région en particulier, sur un nombre restreint de masses d'eaux ou sur un nombre limité de composés, comme les recherches de Berryman et al. (2017) qui ont analysé une cinquantaine de TrOCs de différentes classes dans 9 rivières d'une région restreinte du Québec, de Basiuk et al. (2017) qui ont cherché 14 contaminants dans 4 rivières en Alberta, et de Glozier et al. (2012) qui ont étudié 16 pesticides dans 16 rivières à travers le Canada. Quant aux recherches s'intéressant aux lacs, elles se préoccupent majoritairement de lacs de plus de 1 000 km^2 (Evans and Muir, 2016; Law et al., 2006), en particulier dans la région des Grands Lacs, qui font l'objet de suivi de plusieurs composés persistants, bioaccumulables et toxiques depuis les années 1970 (Gewurtz et al., 2011). Le lac Ontario est le Grand Lac le plus étudié, avec des recherches portant sur l'analyse de POPs (X. Zhang et al., 2020), de composés perfluorés (Myers et al., 2012), de différents PPCPs (Li et al., 2010) ou spécifiquement d'antibiotiques (Nakata et al., 2005) dans ce lac uniquement, ou conjointement avec d'autres Grands Lacs pour l'analyse de composés pharmaceutiques (Metcalfe et al., 2003), de POPs (Marvin et al., 2004) ou de pesticides (Struger et al., 2004).

Des études locales sur des lacs de plus petite taille ont également été réalisées. Dès 1988, des pesticides, maintenant réglementés, ont été étudiés et retrouvés dans des lacs du sud du Labrador (Lockerbie and Clair, 1988). Depuis, plusieurs études sur des POPs ont été effectuées, notamment dans des lacs arctiques (Cabrerizo et al., 2019; Diamond et al., 2005; Helm et al., 2002) et dans un lac de haute altitude de l'ouest canadien (Blais et al., 2001). Les régions arctique et subarctique ont suscité l'intérêt pour l'analyse d'une plus grande variété de TrOCs (34 et 15, respectivement) dans l'étude de 7 lacs du Nunavut (Stroski et al., 2020) et de 10 lacs des Territoires du Nord-Ouest (Liu et al., 2022). Les lacs de la région des sables bitumineux ont aussi été plusieurs fois analysés, particulièrement pour des composés polyaromatiques (Arciszewski et al., 2022; Kelly et al., 2009). La présence de 25 à 44 pesticides a été étudiée dans 10 lacs de l'Ontario (Kurt-Karakus et al., 2011; Murray et al., 2011), tandis que 48 TrOCs de différentes classes ont été recherchés dans 7 lacs ontariens servant de sources d'eau potable (Kleywegt et al., 2011). Des composés perfluorés ont quant à eux été analysés dans 4 lacs des Rocheuses (Loewen et al., 2008) et dans un lac d'assez grande taille de la zone de Grands Lacs (Helm et al., 2011). Ce dernier a également été l'objet d'une étude de l'utilisation de l'analyse de la caféine comme marqueur de rejets domestiques où elle a été retrouvée jusqu'à 77 ng/L (Kurissery et al., 2012). Plusieurs composés pharmaceutiques ont été observés dans le Lac Saint-Charles au Québec à des concentrations allant jusqu'à 1 µg/L pour l'acide salicylique (Pulicharla et al., 2022). Il n'existe toutefois pas d'informations homogènes disponibles sur les lacs de l'ensemble du territoire, en particulier pour ceux de plus petite taille malgré leur nombre et leur importance locale et générale.

Bien que faibles, les concentrations de TrOCs observées dans les eaux naturelles peuvent suffire à engendrer des effets néfastes sur des organismes vivants.

1.2.2. Écotoxicité et effets sous-létaux

Certains TrOCs sont reconnus comme dangereux pour l'humain, pour d'autres espèces animales ou pour des espèces végétales (Huang et al., 2021; Wilkinson et al., 2016). Ce danger est évalué en laboratoire par des tests de toxicité réalisés sur des cibles moléculaires spécifiques pour les humains ou sur des espèces modèles des règnes animal et végétal.

Pour la toxicité humaine, il existe des tests *in vitro* visant des cibles moléculaires liées à des troubles spécifiques. Les principaux impacts recherchés sont les potentiels de carcinogénicité, de mutagénicité et de perturbation endocrinienne (Kojima et al., 2013). Différents impacts sont attendus en fonction des classes de contaminants. Un impact très étudié récemment est la perturbation endocrinienne. Certains composés ont une structure proche d'hormones naturelles et vont agir sur les cibles habituelles de ces molécules, modifiant ainsi la régulation des hormones et pouvant mener par exemple à des troubles de la fertilité.

Cependant, les TrOCs peuvent également affecter les organismes vivant dans les milieux contaminés. Il est donc nécessaire d'évaluer leur écotoxicité. Pour cela, des tests *in vivo* sont effectués sur des espèces modèles sélectionnées en fonction du milieu d'intérêt. Pour la contamination de l'eau, le choix se porte principalement sur des espèces de poisson, de zooplancton et d'algues, qui représentent différents niveaux de la chaîne trophique des eaux naturelles (Mesquita et al., 2023). Afin d'étudier la toxicité des contaminants, ces espèces modèles sont exposées à différentes concentrations de composés pouvant être trouvés dans les écosystèmes aquatiques et différents critères d'évaluation sont observés. Le premier critère évalué dans les études écotoxicologiques est la toxicité aigüe ou chronique, qui est mesurée par la concentration létale médiane (CL_{50}) ou la concentration effective médiane (CE_{50}). Ces seuils correspondent à la concentration à laquelle on observe un effet pour 50% des individus tests, soit la mort pour la CL_{50} ou un autre effet observable pour la CE_{50} comme l'inhibition de la croissance. Le temps d'exposition permettant de mesurer la toxicité aigüe ou chronique varie selon les espèces testées. Par exemple, pour évaluer la toxicité aigüe d'un composé, l'exposition sera habituellement de 96h pour les tests sur les poissons, alors qu'elle sera de 48h pour l'espèce de zooplancton *Daphnia magna*.

Des concentrations proches de celles mesurées dans l'environnement ont démontré une toxicité pour les organismes aquatiques pour certains composés anthropogéniques (Muniz et al., 2023; Srain et al., 2021). Des tests de toxicité aigüe de différents anti-parasitiques sur l'espèce *D. magna* ont révélé des CE_{50} de 250 ng/L pour l'abamectine (Tišler and Kožuh Eržen, 2006) et de 5.7 ng/L pour l'ivermectine (Garric et al., 2007).

Les tests de toxicité sont le plus souvent effectués pour un seul TrOC bien que, dans l'environnement, ces contaminants se retrouvent en mélanges souvent complexes. Or, il est possible que des interactions entre les composés modifient la toxicité de chacun. Ces contaminants pourraient avoir un effet simplement additif, ou bien un effet synergique ou antagoniste (Leeuwen and Vermeire, 2007). La synergie mène à une toxicité plus élevée que la simple addition, par exemple par l'action sur des récepteurs différents ayant un effet similaire (Cedergreen, 2014). L'antagonisme, à l'inverse, donne une toxicité plus faible que celle attendue par l'addition des effets individuels. Cela pourrait être dû à des mécanismes d'action s'inactivant entre eux, par exemple du fait de la compétition pour une même cible ou en agissant sur des cibles ayant des actions opposées (Y. Zhang et al., 2020). Cependant, il est plus facile d'évaluer la toxicité de chaque composé séparément. En effet, comme chaque écosystème contient un mélange différent de contaminants, il n'est pas possible de tester la toxicité de chaque mélange existant.

Néanmoins, la toxicité d'un composé n'est pas le seul critère d'intérêt en écotoxicologie. En effet, les contaminants peuvent aussi avoir des impacts sur des fonctions importantes pour le maintien des espèces. On appelle ces impacts des effets sous-létaux. Les concentrations pouvant induire des effets sous-létaux sont plus faible que les CL_{50} et CE_{50} et sont souvent plus proches des valeurs environnementales. Les TrOCs peuvent avoir des impacts variés selon leurs structures.

Des effets sur l'expression de gènes impliqués dans plusieurs processus biologiques importants ont par exemple été montrés à des concentrations environnementales allant de 1 à 100 ng/L pour un insecticide à large spectre, le fipronil, sur les larves de *Chironomus riparius*, un insecte dont la phase larvaire est aquatique (Pinto et al., 2024). Cependant, des effets sous-létaux sur des organismes non ciblés par les composés ont également été observés. Les larves de *C. riparius* sont également sensibles à la présence d'additifs industriels tels que le bisphénol A qui induit une augmentation de l'activité de certaines enzymes dès une concentration de 1 ng/L (Lee and Choi, 2007). Une étude récente a démontré une augmentation anormale de la mobilité de *D. magna* à la suite d'une exposition à des concentrations de 100 ng/L de l'antidépresseur fluoxétine (Bedrossiantz et al., 2020). Des concentrations inférieures au ng/L jusqu'à quelques ng/L ont montré un effet de chimioattraction, c'est-à-dire un déplacement vers une substance chimique, pour l'espèce de cilié *Tetrahymena pyriformis* de plusieurs composés pharmaceutiques dont les antibiotiques lincomycine et sulfaméthoxazole et l'analgésique naproxène (Láng and Kőhidai, 2012).

Malgré l'intérêt porté à l'évaluation de l'impact des contaminants, il y a un manque de données expérimentales pour de nombreux composés, en particulier pour les CIE, plus récemment développés ou retrouvés dans l'environnement. Pour pallier ce manque, des outils de prédiction ont été développés, dont les méthodes *in silico* basées sur la relation entre la structure des molécules et leur activité, les QSAR (de l'anglais *Quantitative Structure–Activity Relationship*). Ce sont des modèles mathématiques utilisant des valeurs numériques exprimant les propriétés physico-chimiques des composés comme les descripteurs moléculaires (Voigt and Jaeger, 2023). Ces descripteurs incluent par exemple le poids moléculaire, le coefficient de partition octanol-eau ou le nombre d'anneaux benzéniques (Martin, 2020). Les descripteurs moléculaires de chaque composé sont comparés à des bases de données afin d'estimer notamment leur toxicité. La base de données écotoxicologiques la plus utilisée est ECOTOX (https://cfpub.epa.gov/ecotox/), créée par l'US EPA. Les premiers QSAR ont été développés dans les années 1970 et donnaient déjà des prédictions de toxicité acceptables, comme cela a été démontré pour l'estimation de CL_{50} de près de 50 polluants environnementaux pour des poissons juvéniles de l'espèce *Poecilia reticulate* (Könemann, 1980). Les principaux outils basés sur des modèles QSAR utilisés aujourd'hui sont deux applications développées par l'US EPA, ECOSAR (https://www.epa.gov/tsca-screening-tools/ecological-structure-activity-relationships-ecosar-predictive-model) et T.E.S.T (de l'anglais *Toxicity Estimation Software Tool*, https://www.epa.gov/comptox-tools/toxicity-estimation-software-tool-test), et le logiciel VEGA (https://www.vegahub.eu/portfolio-item/vega-qsar/). Malheureusement, les prédictions ne fonctionnent pas pour les composés hors de leur domaine d'applicabilité, en particulier en ce qui concerne le coefficient de partage octanol-eau (log K_{OW}), et pour les composés dans le domaine d'applicabilité, les valeurs de toxicité prédites contiennent tout de même des erreurs et peuvent ne pas toujours s'approcher de celles mesurées en laboratoire lorsqu'elles existent. L'outil T.E.S.T., sélectionné ici pour obtenir les valeurs de toxicité, présente par exemple des limites dans la gamme de solubilité donnant des valeurs fiables, ce qui est compensé par une alerte lorsqu'un composé est hors du domaine d'applicabilité. De plus, contrairement aux évaluations de risque classiques, les valeurs pour le plus bas niveau trophique sont données pour un microorganisme cilié plutôt que pour une algue, bien que cette espèce soit également un modèle très utilisé en écotoxicologie. Enfin, des valeurs ne sont pas toujours produites, ce qui peut mener à une estimation incomplète de l'impact de la contamination organique trace sur les organismes aquatiques. Cependant, les prédictions donnent des valeurs adéquates pour une grande gamme de contaminants, et les modèles QSAR continuent à être développés et améliorés. Ces modèles permettent ainsi d'estimer des valeurs seuils dans l'attente de données expérimentales. Leur utilisation est donc nécessaire dans un contexte où le développement de nouveau composés est en constante augmentation et ne permet pas la mesure immédiate des valeurs de toxicité.

Afin de comprendre le potentiel impact des TrOCs sur les écosystèmes aquatiques, il est nécessaire d'effectuer une évaluation du risque environnemental. En effet, le risque encouru par les espèces aquatiques dépend de multiples facteurs tels que la concentration et la persistance des contaminants, leur biodisponibilité ainsi que leur bioaccumulation et leur bioamplification dans la chaîne trophique, mais aussi le cycle de vie et le stade de développement des organismes, tout comme des caractéristiques propres au milieu pouvant affecter les propriétés des TrOCs. Une approche simplifiée permettant d'estimer ce risque potentiel est de prendre en compte non seulement le danger posé par les contaminants, mais aussi l'exposition des espèces à ces contaminants. L'évaluation du risque telles que définie par les directive et règlements de la Commission Européenne consiste alors à mettre en relation les concentrations environnementales aux valeurs de toxicité obtenues expérimentalement ou estimées (Commission Européenne, 1994, 1993; Conseil Européen, 1993). Afin d'avoir des valeurs conservatrices du risque posé par un composé, ses concentrations sont comparées à la concentration sans effet prédite (PNEC, de l'anglais *predicted no effect concentration*), qui est la concentration en dessous de laquelle aucun effet n'est attendu (Chhipi-Shrestha et al., 2022). La PNEC est le plus souvent calculée en divisant la plus basse valeur de toxicité disponible par un facteur d'évaluation. Lors de l'utilisation de données de toxicité aigüe, ce facteur est de 1000 afin de prendre en compte les incertitudes quant au réel impact du composé dans l'environnement (Leeuwen and Vermeire, 2007). Le ratio entre la concentration environnementale d'un contaminant et sa PNEC, appelé quotient de risque (QR), est alors utilisé pour estimer le risque d'apparition d'effets indésirables dans l'environnement. Plus ce quotient est élevé, plus les chances d'observer des effets est grand. Les QR suivent habituellement la classification suivante : un risque faible pour des QR inférieures 0,1, un risque modéré entre 0,1 et 1, un risque élevé entre 1 et 10 et un risque très élevé au-dessus de 10 (Zhang et al., 2023).

Dans le cadre de l'analyse du risque environnemental de mélanges de contaminants, il peut être difficile de prendre en compte les interactions dans l'effet des différents composés. En effet, les mélanges complexes retrouvés dans l'environnement peuvent inclure des composés ayant le même mode d'action, qui peuvent parfois avoir des effets synergiques, et des composés aux modes d'action différents, dont les effets peuvent être synergiques, antagonistes ou simplement additifs. Cependant, les concentrations environnementales sont le plus souvent de l'ordre du ng/L au µg/L, or les interactions sont observées à des concentrations plus élevées, proche du mg/L et au-delà (Boobis et al., 2011; Cedergreen, 2014). Il est donc admis que les modèles additifs sont suffisants pour estimer l'impact des mélanges environnementaux de TrOCs (Escher et al., 2020). L'addition des QR des composés individuels est d'ailleurs la méthode privilégiée dans les études d'analyse de risque environnemental ou humain à travers le monde, tant en Asie (Pei et al., 2022; Yan et al., 2014) qu'en Europe (Boberg et al., 2019; Ginebreda et al., 2010).

Plusieurs évaluations du risque environnemental ont été effectuées pour des lacs à travers le monde afin d'estimer l'impact de leur contamination sur les organismes aquatiques. En Chine, des études ont analysé le risque de plusieurs TrOCs dans un lac chacune. Zhang et al. (2018) ont obtenu des QR ne dépassant pas le risque faible dans le lac Baiyangdian, avec un maximum de 0,08 pour l'antibiotique vétérinaire tylosine. Des risques plus élevés ont été estimés pour des TrOCs et des métaux observés dans le lac Tai, avec un QR individuel allant jusqu'à 1,18 pour le nickel et une somme des QR de 4,2 pour l'ensemble des composés (Pei et al., 2022). Pour le lac Taihu, des QR très élevés ont été calculés, notamment pour la présence d'hormones dans le lac donnant des QR jusqu'à 18 et une somme de 67 (Yan et al., 2014). Des QR modérés ont été mesurés en Inde dans l'étude de quelques PPCPs dans 6 lacs urbains avec un quotient maximal de 0,43 pour l'antibiotique humain ciprofloxacine (Archana et al., 2017).

De telles recherches ont aussi pris place en Europe, notamment en Hongrie, dans le plus grand lac peu profond d'Europe, le lac Balaton, et dans un lac urbain en Grèce, le lac Pamvotis. Les organismes aquatiques du lac Pamvotis semblent à risque dû à la détection de l'antibiotique d'utilisation mixte érythromycine donnant des QR jusqu'à 7 (Nannou et al., 2015). Quant au lac Balaton, plusieurs TrOCs y ont été retrouvés à des concentrations pouvant poser un risque élevé tels que la caféine (QR max 1,16), les hormones estrone et estradiol (QR max 5,52 et 9,80, respectivement), et le diclofénac qui a donné un QR très élevé de 40 (Molnar et al., 2021). L'étude à plus grande échelle d'Anagnostopoulpou et al. (2022) sur la contamination de 18 lacs à travers la Grèce a également porté sur l'évaluation du risque environnemental encouru par ces lacs. Des plus de 470 TrOCs analysés, seul le fongicide carbendazime a donné un risque élevé (QR $\geq$ 1) pour les daphnies, tandis que des risques modérés ($0{,}1 \leq$ QR < 1) ont été calculés pour le chasse-moustiques DEET pour les daphnies et pour l'analgésique ibuprofène pour les algues.

En Amérique du Nord, une analyse de risque de la présence de 54 PPCPs a été effectuée dans le lac Michigan (Blair et al., 2013). Cette étude a démontré un risque élevé pour la caféine et son métabolite principal la paraxanthine (max QR 5,9 et 6,9 respectivement) et pour l'antibiotique ofloxacine (max QR 1,2) à 1,6 km au large d'un rejet de STEP, et d'un risque modéré à 3,2 km du rejet pour l'antibiotique humain sulfaméthoxazole et l'analgésique codéine. Aux États-Unis, 6 lacs d'une même chaîne ont été étudiés pour 110 TrOCs et le risque le plus élevé a été démontré pour l'herbicide atrazine (Pronschinske et al., 2023). Au Canada, une analyse de risque a été effectuée pour 7 composés pharmaceutiques détectés dans 7 lacs arctiques, menant à un calcul de risque élevé pour le sulfaméthoxazole pour l'algue *Pseudokirchneriella subcapitata* (QR 2,5) (Stroski et al., 2020). Néanmoins, il n'existe pas d'étude à grande échelle du risque environnemental encouru par les organismes lacustres.

Effectuer une évaluation des risques environnementaux permet d'avoir une meilleure idée des potentiels impacts de la contamination observée dans l'environnement. Cependant, afin de limiter cette contamination, il est important de connaître les facteurs qui y mènent.

1.3. Principaux facteurs influençant la contamination des lacs

Mieux caractériser les facteurs influençant la contamination en TrOCs des lacs permettrait de prioriser les efforts de réduction de cette contamination en mettant l'accent sur les sources les plus importantes ou les modes d'entrées dans l'environnement pouvant être limités.

1.3.1. Utilisation du sol et activités anthropiques

Les TrOCs trouvés dans les eaux de surface peuvent provenir du rejet direct de contaminants dans les cours d'eau, mais aussi du ruissellement dans leurs bassins versants mène au transfert vers l'environnement aquatique de contaminants présents dans les sols (Fairbairn et al., 2015; Guzzella et al., 2018). Les sources de TrOCs peuvent être ponctuelles ou diffuses (**Figure 3**). Parmi les principales sources ponctuelles, il y a les activités récréatives prenant place directement sur les lacs (e.g., baignade, pêche, navigation de plaisance) qui se déroulent le plus souvent en été et qui peuvent être des sources de contaminants spécifiques tels que les filtres à ultraviolets (UV) utilisés dans les crèmes solaires ou les composés utilisés comme chasse-moustiques tels que le DEET (Aronson et al., 2012; Langford and Thomas, 2008). Les effluents de STEP ainsi que les débordements d'égouts et de systèmes d'épuration individuels (i.e., les fosses septiques) déversés dans les cours d'eau tributaires peuvent être la source de différentes familles de contaminants dont les PPCPs (Guyader et al., 2018; Subedi et al., 2015), tandis que les rejets d'industries peuvent libérer des additifs tels que des plastifiants ou des retardateurs de flammes (Chokwe et al., 2020). L'utilisation de composés tels que des antibiotiques et des hormones en aquaculture sont également une source ponctuelle de TrOCs pour les lacs (Kolodziej et al., 2004; Lai et al., 2018). Quant aux sources diffuses, elles sont principalement liées à une utilisation de composés organiques sur des surfaces plus vastes liées à différentes utilisations du sol. Elles entraînent une contamination des eaux de surfaces par ruissellement sur ces surfaces et par infiltration (Müller et al., 1998). Une première source diffuse est l'épandage de boues d'épuration sur les terres agricoles, qui peut apporter des résidus semblables à ceux retrouvés dans les rejets de STEP (Fijalkowski, 2019; Ivanová et al., 2018). L'utilisation de produits phytosanitaires en agriculture ou dans le milieu urbain, sur les pelouses ou les terrains de golf par exemple, peut mener à la présence de pesticides dans les lacs (Metcalfe et al., 2016; Wilson, 2013). L'urbanisation est également source d'autres catégories de contaminants par le lessivage de décharges par exemple, qui peuvent libérer des composés variés (Masoner et al., 2016; Pozo et al., 2023), ou par l'émission de contaminants tels que des hydrocarbures aromatiques polycycliques ou des phtalates par la circulation routière (Järlskog et al., 2021).

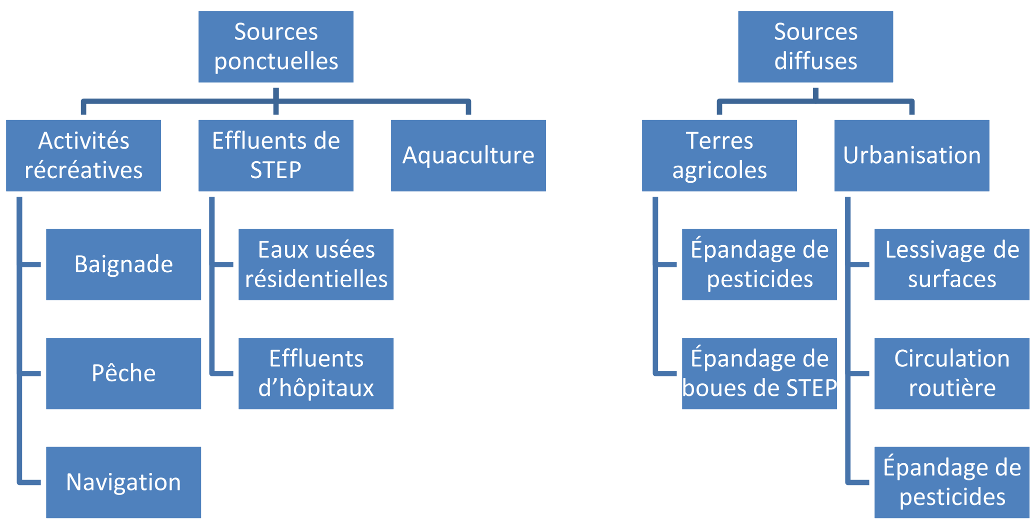

Figure 3. Principales sources de contamination anthropique.

L'importance de ces différentes sources a été évaluée par plusieurs études à travers le monde. Néanmoins, elles s'intéressent principalement à des cours d'eau et non à des lacs (Meyer et al., 2019). Cependant, les facteurs affectant les lacs peuvent être similaires étant donnée l'interconnectivité des systèmes aquatiques. En effet, les cours d'eau et les eaux souterraines sont les apports principaux des lacs.

Les études de sources de contamination organique dans les lacs ont principalement été effectuées en Chine, souvent sur un lac et son bassin versant plutôt que sur plusieurs lacs. Ces études ont démontré l'importance des activités humaines sur la détection de différentes classes de contaminants tels que des pesticides, des PPCPs et des plastifiants par des apports diffus, comme l'agriculture, l'urbanisation et les émissions atmosphériques, ou des apports ponctuels, tels que les effluents de STEP, d'industries et d'eaux usées non traitées et l'aquaculture (An et al., 2023, 2022; Bhutto et al., 2021; Chen et al., 2022; Duan et al., 2021; He et al., 2021; Huang et al., 2022; Liu et al., 2023). Une étude portant sur l'analyse de pesticides dans 14 lacs de la région du Wuhan en Chine a mis en avant l'importance de l'urbanisation, de l'aquaculture et de la présence de bateau de pêche dans la contamination des lacs (Cui et al., 2017). Dans les Philippines, l'étude de 3 lacs voisins a montré l'impact des activités humaines proches des lacs, tels que l'aquaculture, les rejets d'eau usée non traitée et le tourisme, menant à des profils de contamination différents dans ces 3 lacs malgré leur proximité (Dimzon et al., 2018). En Europe, l'étude d'un lac alpin en Suisse et de 4 lacs isolés en Irlande ont confirmé l'apport de TrOCs par les dépositions atmosphériques provenant notamment de STEP (Aherne et al., 2023; Müller et al., 2011). L'analyse d'édulcorants de synthèse dans 3 lacs finlandais a quant à elle illustré l'influence des rejets de STEP et de fosses septiques ainsi que de potentielles fuites de stations de pompage d'eaux usées sur la contamination des eaux de surface (Perkola and Sainio, 2014). L'impact des fosses septiques a également été prouvé dans l'étude de 5 lacs aux États-Unis recevant des rejets de systèmes d'épuration individuels, bien que des sources agricoles et récréatives aient également été soupçonnées (Guyader et al., 2018). Au Canada, 2 études ont porté sur les sources de contamination du Lac Ontario. L'analyse de PPCPs dans 2 zones littorales du lac a démontré l'impact des rejets de STEP, mais aussi du degré d'urbanisation et de la densité de population dans le littoral du lac (Helm et al., 2012). L'étude de la présence de pesticides dans plusieurs zones du Lac Ontario a également montré l'importance de sources urbaines telles que les rejets de STEP et le ruissellement sur les terrains de golf en plus des sources agricoles (Metcalfe et al., 2016). Ces analyses portent donc généralement sur un système uniquement, tandis que les études sur un grand nombre de bassins versants indépendants sont rares.

Néanmoins, ces études exploratoires permettent d'émettre des hypothèses sur l'importance des différents facteurs sur la contamination des eaux de surfaces. Cependant, ces hypothèses doivent ensuite être testées afin de confirmer la contribution de chaque facteur à l'ampleur de la contamination. Pour correctement évaluer l'impact des activités anthropiques sur la contamination des lacs, il est nécessaire de prendre en compte les facteurs environnementaux pouvant moduler leurs effets.

1.3.2. Facteurs environnementaux

Des facteurs biotiques et abiotiques peuvent influencer les niveaux de contamination des lacs. Le principal processus biotique impliqué est la biodégradation (Berkowitz et al., 2014). En effet, bien que certains contaminants organiques soient récalcitrants à la dégradation par les microorganismes, c'est un processus non négligeable d'atténuation naturelle de la contamination des eaux de surface (Blunt et al., 2018). Les concentrations de TrOCs dans la phase dissoute peuvent également être réduites par sorption sur les biofilms, formés de microorganismes associés à des substances extracellulaires (Wang et al., 2019).

Les principaux processus abiotiques entrant en jeu dans le devenir des TrOCs sont des caractéristiques propres aux lacs, des composantes de leurs bassins versants ou des facteurs climatiques qui peuvent influencer les niveaux de contamination dans ces écosystèmes (Schimmelpfennig et al., 2016). Ces différents processus sont détaillés ci-dessous.

Concernant les caractéristiques des lacs, le temps de résidence, qui définit le temps entre l'entrée d'une masse d'eau dans le lac et sa sortie, peut influer sur les concentrations de TrOCs car une plus grande rétention de l'eau peut mener à une accumulation des contaminants dans le lac (Czekalski et al., 2015). La présence de matière organique dissoute joue aussi un rôle important dans la concentration des TrOCs en phase dissoute. En effet, les contaminants peuvent se lier à la matière organique par sorption (Hung et al., 2006), ce qui pourrait augmenter la sédimentation des TrOCs présents dans l'épilimnion et augmenter leurs concentrations dans l'hypolimnion en cas de désorption. D'autres facteurs peuvent impacter la dilution des TrOCs, comme le volume du lac (Smith et al., 1997) ainsi que le rapport entre la surface d'un lac et celle de son bassin versant (Mills et al., 2022). En effet, pour un bassin versant similaire et des sources semblables, un petit lac aura une concentration de contaminants plus élevée qu'un plus grand lac qui aurait alors un plus grand facteur de dilution. La topographie est une autre composante du bassin versant pouvant déterminer le volume de TrOCs cheminant vers les lacs. En effet, la pente du bassin versant peut favoriser le transport des contaminants et ainsi faciliter leur entrée dans les eaux de surface (Garcia et al., 2015).

Au niveau climatique, les principaux facteurs pouvant impacter les niveaux de contamination dans l'eau des lacs sont : les précipitations, qui mobilisent les TrOCs et les introduisent dans les écosystèmes aquatiques en favorisant le ruissellement (Kim and Kannan, 2007; Miao et al., 2020) ; la température, qui peut favoriser la dégradation des composés par hydrolyse (Berkowitz et al., 2014) ; et l'irradiation solaire, qui est une mesure d'énergie solaire par unité de surface et dont l'augmentation amplifie la photolyse des composés dans l'épilimnion (Berkowitz et al., 2014; Jasper and Sedlak, 2013).

De plus, ces différents facteurs peuvent interagir entre eux, de façon synergique ou antagoniste. Leurs impacts peuvent donc parfois se conjuguer, comme la présence de matière organique dissoute qui, en plus de diminuer les concentrations de TrOCs par sorption, peut décupler leur biodégradation et ainsi réduire encore leurs concentrations (Tang et al., 2017). La biodégradation est également favorisée par les températures élevées (Jasper et al., 2014). La pente et les précipitations peuvent aussi avoir un effet combiné sur l'augmentation du ruissellement dans les bassins versants qui peut être plus grand que la somme de leurs effets individuels (Li et al., 2020), ce qui peut mener à une plus grande contamination.

Cependant, l'augmentation du ruissellement, peut également mener à une plus grande concentration de matière organique dissoute qui favorise l'atténuation naturelle des TrOCs (Queimaliños et al., 2019). De plus, une forte pente est généralement liée à une plus grande profondeur du lacs, donc à un volume plus important qui pourrait compenser l'apport de contaminants par dilution. Néanmoins, la photodégradation des contaminants est favorisée par une plus faible profondeur de lac (Schimmelpfennig et al., 2016), ce qui pourrait contrebalancer en partie la faible dilution dans ces lacs.

Les lacs sont des écosystèmes complexes qui sont influencés par de nombreux facteurs tant naturels qu'anthropiques. Comprendre leur fonctionnement est donc un enjeu de taille mais nécessaire pour améliorer leur gestion et réduire leur potentielle contamination.

1.4. Objectifs du projet

Les lacs jouent un rôle important, tant pour l'environnement que pour les services écosystémiques rendus aux sociétés humaines, en particulier au Canada. Cependant, ces écosystèmes sont sensibles aux activités anthropiques, menant notamment à une contamination chimique pouvant déstabiliser l'équilibre de ces lacs par leurs impacts sur les organismes lacustres. En particulier, les contaminants organiques à l'état de trace sont omniprésents dans l'environnement, notamment dans les écosystèmes aquatiques. Or, ces contaminations et leurs impacts sur les lacs sont encore peu évalués à grande échelle. De plus, les facteurs humains liés à la présence et à l'ampleur de cette contamination ont principalement été étudiés dans les rivières. Et, bien que des corrélations aient été mises en évidence, il y a un manque de recherche de causalité entre ces facteurs et les contaminations observées. Mieux caractériser l'impact des activités humaines sur les lacs permettrait de guider nos efforts dans la réduction de la contamination et ainsi d'améliorer la qualité de ces écosystèmes. Mon projet de recherche vise donc à évaluer la présence de ces contaminants dans les lacs canadiens, leurs potentiels impacts sur le biote lacustre ainsi que les facteurs menant à cette contamination.

Le premier objectif de cette thèse est de caractériser la contamination organique trace des lacs à l'échelle du Canada et d'évaluer les risques que les contaminants retrouvés pourraient poser pour les organismes aquatiques. Pour cela, une méthode d'analyse multi-résidus comprenant une étape de concentration par extraction en phase solide suivie d'une séparation par chromatographie liquide couplée à une détection par spectrométrie de masse en tandem a été développée pour 54 TrOCs représentatifs de diverses activités humaines. Ces contaminants ont ensuite été analysés dans l'eau de 290 lacs représentatifs de la diversité des lacs canadiens de moins de 100 km^2, échantillonnés à travers le pays. Dans un deuxième temps, une évaluation du risque environnemental encouru par les écosystèmes lacustres échantillonnés a été effectuée afin de mettre en perspective les concentrations de TrOCs retrouvées. Pour cela, les résultats de l'analyse des contaminants ont été mis en relation avec les valeurs seuils de toxicité pour les 3 espèces aquatiques de différents niveaux trophiques incluses dans l'outil T.E.S.T. de l'US EPA. Les travaux en lien avec cet objectif sont présentés sous forme d'article au chapitre 2.

Étant donné l'impact potentiel des TrOCs observés dans les lacs canadiens, comme mentionné dans le chapitre 2, il est crucial d'explorer des moyens de diminuer les niveaux de contamination. Or, pour réduire efficacement la présence de ces TrOCs et leurs impacts sur les lacs, il est nécessaire de mieux comprendre les facteurs qui mènent à cette contamination afin de diriger les efforts de réduction vers les sources les plus importantes. C'est pourquoi le second objectif de cette thèse est de spécifier les relations entre de potentiels facteurs anthropiques, tels que l'utilisation du sol et la présence de sources ponctuelles, et l'ampleur de la contamination des lacs canadiens. Des hypothèses basées sur les résultats d'études exploratoires ont donc été testées afin de valider l'impact de potentielles sources de contamination ponctuelles et diffuses. Pour cela, des modèles de régression linéaire et logistiques ont été développés pour valider les hypothèses. Un modèle prédictif a également été établi afin d'estimer les concentrations de pesticides dans l'eau des plus de 200 000 lacs de moins de 100 km^2 au Canada. La recherche effectuée pour cet objectif est présentée sous forme d'article au chapitre 3.

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CHAPITRE 2. LES CONTAMINANTS ORGANIQUES À L'ÉTAT DE TRACES DANS LES EAUX DE LAC : PRÉSENCE ET ÉVALUATION DU RISQUE ENVIRONNEMENTAL À L'ÉCHELLE NATIONALE AU CANADA

2.1. Notes préliminaires

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Des modifications ont été effectuées par rapport à la version soumise selon les commentaires du jury.

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2.3. Contribution des auteurs

L.L., Y.H. et P.A.S. ont conçu le plan expérimental. L.L. a participé à l'échantillonnage à grande échelle des lacs canadiens, a développé la méthode d'analyse, a réalisé l'ensemble des expériences et l'analyse de données, et a rédigé le manuscrit. L.L., H.C., Y.H. et P.A.S. ont révisé le manuscrit. Tous les auteurs ont contribué à cet article et ont approuvé la version finale.

2.4. Abstract

Numerous contaminants are produced and used daily, a significant fraction ultimately finding their way into natural waters. However, data on their distribution in lakes is lacking. To address this gap, the presence of 54 trace organic contaminants (TrOCs), representative of various human activities, was investigated in the surface water of 290 lakes across Canada. These lakes ranged from remote to highly impacted by human activities. In 88% of the sampled lakes, contaminants were detected, with up to 28 detections in a single lake. The compounds most frequently encountered were atrazine, cotinine, and deethylatrazine, each of which was present in more than a third of the lakes. The range of detected concentrations was from 0.23 ng/L to about 2 200 ng/L for individual compounds, while the maximum cumulative concentration exceeded 8 100 ng/L in a single lake. A risk assessment based on effect concentrations for three aquatic species (*Pimephales promelas*, *Daphnia magna*, and *Tetrahymena pyriformis*) was conducted, revealing that 6% of lakes exhibited a high potential risk for at least one species. In 59% of lakes, some contaminants with potential sub-lethal effects were detected, with the detection of up to 17 TrOCs with potential impacts. The results of this work provide the first reference point for monitoring the evolution of contamination in Canadian lakes by TrOCs. They demonstrate that a high proportion of the sampled lakes bear an environmentally relevant anthropogenic chemical footprint.

Keywords: multi-residue analysis, pharmaceuticals, pesticides, ecological impacts, large-scale

2.5. Introduction

Many organic chemicals are produced and used daily around the globe. Some of these compounds are employed as pesticides in agriculture, some constitute the pharmaceutically active ingredients of common drugs, while others are used as consumer product additives such as plasticizers or flame retardants (Fischer et al., 2017; Sousa et al., 2018). These compounds are widely used and can find their way into water bodies mainly through surface runoff or municipal and industrial wastewater discharge (Pal et al., 2010; Yang et al., 2017). Trace organic contamination from these chemicals can lead to considerable impacts on aquatic ecosystems worldwide.

Water is a most valuable resource, and in Canada, lakes hold a very important place. Indeed, this country is home to more than 2.4 million lakes (Cooke and Murchie, 2015) that are used for hydroelectricity production, as sources for drinking water and irrigation, for fisheries and recreational fishing, as well as for recreational purposes (Inácio et al., 2022; Reynaud and Lanzanova, 2017). These ecosystems also sustain a high biodiversity and are of great importance to Indigenous communities. Moreover, lakes act as sinks of contamination as they integrate what is released in their watersheds, partly through surface runoff (Müller et al., 1998). Nevertheless, large-scale studies on surface water contamination mostly focus on rivers rather than lakes (Bradley et al., 2017; Focazio et al., 2008; Glozier et al., 2012; Kolpin et al., 2002; Loos et al., 2009), while literature on lake contamination is present but usually concerned with the analysis of a limited number of lakes or localized areas (Anagnostopoulpou et al., 2022; Archana et al., 2017; Blair et al., 2013; Ferrey et al., 2015; Liu et al., 2022; Maasz et al., 2019; Malnes et al., 2022; Müller et al., 1997; Sharma and Hanigan, 2021; Sodré et al., 2018). Despite the unique status of lakes in Canada, little attention has been paid to their health status nationwide. Most studies focus on understanding the Great Lakes and other large lakes (e.g., Lake Winnipeg and Great Slave Lake), leaving behind our comprehension of smaller lakes that are numerous and of great local and global significance (Baker et al., 2022; Evans and Muir, 2016; Law et al., 2006; Li et al., 2010; Marvin et al., 2004; Myers et al., 2012). In this context, the NSERC Lake Pulse Network aimed to provide data on lake health across Canada, including the presence of organic contamination (Huot et al., 2019).

Trace organic contaminants (TrOCs) can have ecological consequences even at low concentrations, exhibiting acute and chronic toxicity as well as sub-lethal effects (Anderson et al., 2015; Chaturvedi et al., 2021; Kidd et al., 2007; Petrie et al., 2015; Warren et al., 2021; Wilkinson et al., 2016). Therefore, it is essential to assess the potential impacts of compounds detected in surface water. Although a multiplicity of factors can modulate these impacts, they can be estimated by taking into account a limited number of parameters. Risk assessments can be carried out by comparing quantified concentrations found in the environment to toxicity thresholds such as predicted no effect concentrations (PNECs) (Chhipi-Shrestha et al., 2022; Dong et al., 2022; Molnar et al., 2021; Tang et al., 2015). PNEC values are usually obtained using 50% lethal concentration (LC_{50}) or 50% effect concentration (EC_{50}) of model aquatic organisms divided by an assessment factor, typically of 1 000. Risk quotients (RQs) can then be calculated by dividing environmental concentrations by the corresponding PNEC value, with RQ values of 1 and above indicating high ecological risk (Zhang et al., 2023). Unfortunately, experimental data on the effects of TrOCs on aquatic organisms is fragmentary. To address this gap, in-silico toxicity prediction tools have been developed. These consist of mathematical models that are used to predict toxicity levels based on molecular structure, similarity, or molecular descriptors such as molecular weight or octanol-water partition coefficients (Martin, 2020; Ortiz de García et al., 2013; Voigt and Jaeger, 2023).

Considering this information, this study aimed to address the knowledge gap on trace organic contamination of Canadian lakes and its impacts on aquatic organisms. To achieve this, 290 lakes, representative of the diversity of Canadian lakes smaller than 100 km^2, were sampled across the country. Subsequently, the concentration of a group of 54 TrOCs, which consisted of pharmaceuticals, personal care products, consumer product additives, and pesticides, was determined in those samples. Finally, an environmental risk analysis was conducted to assess the potential impacts of the observed contamination on these valuable ecosystems. This data will serve as a reference point for future monitoring work.

2.6. Experimental methods

2.6.1. Lake selection

The lake selection has been described in depth elsewhere (Huot et al., 2019). Briefly, a stratified random selection of Canadian lakes was done, with ecozone, lake size, and a human impact index used as group stratifications. Ecozones are regions categorized by their ecological, geological, and climatic features. Lake size ranged from 0.1 to 100 km^2, and the human impact index (HII) was calculated from land use data within the lakes' watersheds. To assess the HII, each land-use pixel was assigned a value between 0 and 1 related to the potential impact of its category: 0 for natural landscapes, 0.5 for forest loss and pasture, and 1 for mines, agriculture, and urban land-use. The HII for each lake was determined as the average of each pixel in its watershed. This index was developed to rapidly classify lakes based on land use data from public sources and it was used for group stratification to select lakes with a range of potential human impacts.

This selection method allowed for the sampling of 290 lakes representative of the distribution of Canadian lakes accessible by road or forestry road (**Figure 4**). Information on the sampled lakes can be found in the Annexe 1 **Tableau 6** and additional details can be accessed on the Lake Pulse web portal (https://lakepulse.ca/lakeportal/).

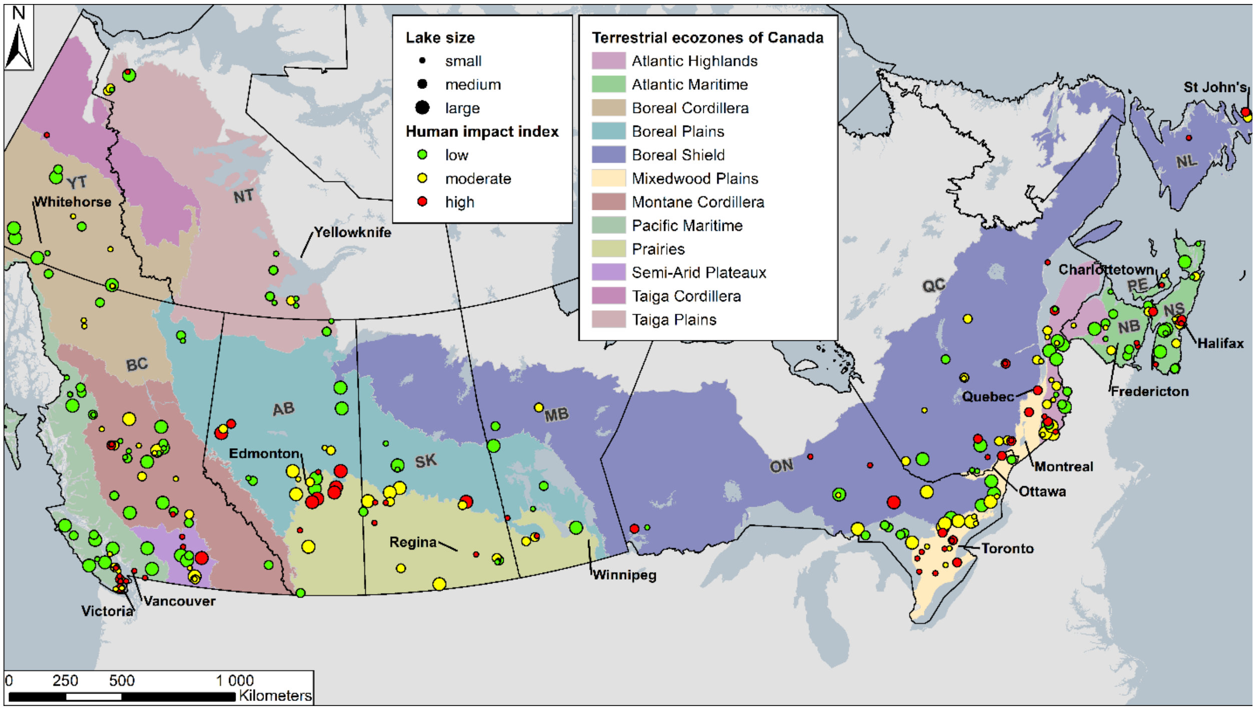

Figure 4. Location of the 290 lakes sampled in the summers of 2017, 2018 and 2019. Canadian provinces and territories: YT: Yukon, NT: Northwest Territories, BC: British Columbia, AB: Alberta, SK: Saskatchewan, MB: Manitoba, ON: Ontario, QC: Quebec, NB: New Brunswick, PE: Prince Edward Island, NS: Nova Scotia, NL: Newfoundland and Labrador.

2.6.2. Sampling methods

Sampling took place between July and September in three consecutive years (2017, 2018 and 2019). A single sub-surface water sample was collected at each lake at above the deepest location of the lake. In 2017, samples were collected using a 2-m long tube sampler integrating the euphotic zone, which is the layer that receives enough light to sustain photosynthesis and is defined as twice the Secchi disk depth. The samples were then transferred to 1 L amber high-density polyethylene (HDPE) bottles after 3 rinses with lake water. In 2018 and 2019, surface water was collected directly in HDPE bottles without integration of the euphotic zone. Samples were then stored at -20°C within 1 hour of collection until analysis to limit degradation of the compounds of interest.

2.6.3. Contaminant selection

Contaminants representative of relevant human activities (e.g., agricultural, urban, industrial, recreative) were chosen for their ubiquitous presence in the environment (Bradley et al., 2017; Focazio et al., 2008; Kolpin et al., 2002), their toxicity (Ortiz de García et al., 2013), and their extensive use in Canadian provinces (Murray et al., 2011). The selection of 54 contaminants comprises 27 pharmaceutical products, 1 artificial sweetener, 3 personal care products, 7 consumer product additives and 16 pesticides (Annexe 1 **Tableau 7**).

2.6.4. Chemicals and Materials

All reagents and analytical standards were purchased as solids or liquids (purity > 92%) from MilliporeSigma (Oakville, Canada), ChemService (West Chester, USA), or MedChemExpress (Monmouth Junction, USA). Deuterated standards were purchased from CDN isotopes (Pointe-Claire, Canada). Details on selected contaminants are presented in Annexe 1 **Tableau 7**.

Methanol (MeOH), water (H_2O), dimethyl sulfoxide (DMSO), acetonitrile (ACN), and isopropanol (IPA) grade Optima for LC/MS were purchased from Fisher Scientific (Ottawa, Canada). Ultrapure water was obtained from a coupled water purification system composed of a Barnstead E-Pure (Barnstead Lab Water Products, USA) followed by an Advantage A10 (MilliporeSigma, Canada).

Concentrated stock solutions were prepared in various solvents depending on analytes' solubility (Table S2), stored at -20°C, and renewed every 3 or 6 months depending on the stability of the compound. A concentrated working solution was prepared every month and a diluted one every week.

Polymeric reverse-phase Strata-X cartridges (200 mg, 6 mL) were purchased from Phenomenex (Torrance, CA, USA). Glass fiber filters (1.2-mm) and mixed cellulose membrane filters (0.45-mm) were purchased from MilliporeSigma.

2.6.5. Laboratory Sample Preparation

Before analysis, samples were thawed at 4°C for 4 days and were then filtered to 0.45-µm to remove particulate matter. The pH of the filtered samples was adjusted to 6.5 and 200 mg/L of ethylenediaminetetraacetic acid disodium salt dihydrate (Na_2EDTA) was added to prevent analytes from complexing metal ions. Samples were processed in triplicate. When the sampled volume was insufficient, only one or two replicates were processed (see Annexe 1 **Tableau 6**). Sub-samples of 200 mL were concentrated by a factor of 500 by solid-phase extraction (SPE). Deuterated internal standards were added at 200 ng/L, and samples were evaporated to dryness. Finally, samples were reconstituted in H_2O-MeOH (80:20, *v/v*) and transferred to 2 mL amber glass vials for analysis. The sample preparation steps are detailed in Annexe 1, section **A1.1**.

2.6.6. Sample Analysis

Details on the analytical apparatus are given in Annexe 1 **A1.2**, and information on the gradients and mass spectrometer conditions can be found in Annexe 1 **Tableau 8** and **Tableau 9**, respectively. In summary, quantification of contaminants was done by liquid chromatography-triple quadrupole mass spectrometry. Ionization was done with an electrospray source used in positive (ESI+) and negative (ESI-) modes. For analysis in ESI+ mode, the mobile phase was composed of solvent A, formic acid 0.1% *v/v* in H_2O, and solvent B, formic acid 0.1% *v/v* in MeOH. For ESI-, solvent A was 1mM ammonium acetate in H_2O, and solvent B was 1mM ammonium acetate in MeOH. Two multiple reaction monitoring (MRM) transitions were monitored for each contaminant. For each analytical run, a matrix-matched internal calibration was done in non-contaminated lake water with the addition of 11 deuterated compounds selected as internal standards throughout the analytical run (Annexe 1 **Tableau 10**). All data was acquired and processed using TargetLynx V4.2 software.

2.6.7. Quality Assurance and Quality Control

Figures of merit of the analytical method, including extraction recovery percentages, limits of detection (LOD) and of quantification (LOQ), and precision, were evaluated several times during the project. The validation steps are detailed in Annexe 1 **A1.3**, and the results are presented in Annexe 1 **Tableau 10** and

Tableau 11. Before and during sample collection, the use of insect repellent, caffeinated products, and tobacco was avoided to limit sample contamination. Field blanks were collected once every 3 to 4 lakes (83 blanks collected) and laboratory blanks were processed for every sample extraction batch to assess the potential contamination of lake samples. Moreover, fortified laboratory spikes were added to each extraction batch to control for SPE recoveries. Instrumental blanks of H_2O-MeOH (80/20, *v/v*) and calibration controls were also analyzed every 10 to 12 samples. Field and laboratory blanks were prepared using ultrapure water, and extraction and calibration controls were prepared with a pristine lake matrix (Lac Gale, QC).

Concentration values were removed from results when they were below the 95th percentile of field blanks for the corresponding compound (see Annexe 1 **Tableau 10**) and carry over was monitored using laboratory blanks (Focazio et al., 2008). Results presented in this article also passed several quality control criteria. To be included, coefficients of determination of the calibration curves had to be above 0.99, and the analytes had to be quantified in at least two replicates. The quality of the measurements was investigated using the Horwitz Ratio (HorRat), calculated as the ratio of observed relative standard deviation (RSD) to predicted RSD (set at 22% as our concentrations were below 120 µg/L) (Horwitz and Albert, 2006). Results were excluded when the replicates had a HorRat > 1.3, and replicates with HorRat < 0.3 were further investigated to ensure these quantifications were not due to an analytical bias (Rivera and Rodríguez, 2014). Although a second qualitative MRM transition for confirmation purposes was monitored besides a quantitative MRM, the absence of the latter was not critical for data suppression, as concentrations would have been biased towards higher values. Moreover, concentrations were considered as estimated when they were between the LOD and LOQ or when a drift in the calibration was highlighted by controls. These values were not included in descriptive statistics. Data extrapolated over the linearity range were set to the highest linearity value for statistics and graphical representations, and their extrapolated concentrations are mentioned where appropriate. Finally, the concentrations were corrected using SPE recoveries.

2.6.8. Environmental Risk Assessment

To evaluate the risks of contaminants in the sampled lakes, risk quotients were calculated using acute toxicity values for three distinct aquatic species available for the chosen tool across the trophic levels (i.e., fish, crustaceans, and ciliates). The U.S. EPA Toxicity Estimation Software Tool (T.E.S.T., version

5.1.2) was employed to obtain both experimental and estimated toxicity data. Experimental data used by T.E.S.T. is obtained from the ECOTOX knowledgebase (https://cfpub.epa.gov/ecotox/). For compounds without any experimental data, T.E.S.T. uses Quantitative Structure Activity Relationship (QSAR) models to estimate acute toxicity values as well as two sublethal effects: developmental toxicity and mutagenicity (Martin, 2020). Developmental toxicity is defined as the potential of a chemical to disturb the natural development of either humans or animals, before and after birth. Mutagenicity is based on the Ames test. It assesses if a compound can induce mutations in the DNA of the bacterial species *Salmonella typhimurium*. Since T.E.S.T. uses several QSAR methodologies to estimate toxicity values, the consensus method, which gives the average of all methods available, was employed as it is recommended by the U.S. EPA because it gives more accurate results.

Individual risk quotients (*rq*) were calculated as follows:

$$rq_{ijk} = \frac{\mathrm{C}_{ij}}{\mathrm{PNEC}_{jk}} \quad [2.1]$$

where C_{ij} is the concentration of compound *j* in lake *i* and $PNEC_{jk}$ is the predicted no-effect concentration for the compound *j* and the species *k* calculated as follows:

$$\mathrm{PNEC}_{jk} = \frac{\mathrm{EC}_{50\,jk}}{1000} \quad [2.2]$$

where $EC_{50\,jk}$ is the median effect concentration for the compound *j* and the species *k*. The endpoint considered for EC_{50} varies between the three tests included. For fish (model species *Pimephales promelas*) and crustaceans (model species *Daphnia magna)*, the median lethal concentration (LC_{50}) is used, while median growth inhibition concentration (IGC_{50}) is used for ciliates (model species *Tetrahymena pyriformis*). PNEC used for calculations per compound are given in Annexe 1 **Tableau 14**.

As the simultaneous presence of multiple TrOCs in the samples is expected, mixture effects must be taken into account. The toxicity of mixtures of TrOCs with diverse modes of action at low concentrations can be estimated using an additive model (Escher et al., 2020). This model assumes that interactions leading to synergistic or antagonistic effects between contaminants can be neglected and that the risk quotients of all compounds in a mixture can be added to estimate the summed risk quotient (RQ) also known as hazard index (Heys et al., 2016). Therefore, the RQ for all compounds found in each lake was calculated as:

$$\mathrm{RQ}_{ik} = \sum_j rq_{ijk} \quad [2.3]$$

where Σ_j indicates the sum of all compounds for one test species. A high environmental risk is expected when RQ ≥ 1 (Anagnostopoulpou et al., 2022).

2.7. Results and discussion

2.7.1. Overall occurrence of TrOCs in Canadian lakes

The first aim of this study was to evaluate the extent of trace organic contamination in Canadian lakes. The results show that at least one contaminant was found in 88% of the 290 sampled lakes and at least two were detected in 72% of the lakes (**Figure 5**). A high overall detection frequency of one or more contaminants was expected as most of the studied lakes' watersheds support human activities. However, the frequency exceeded expected values as a significant proportion of lakes with low or very low human activity within their watershed were included in the study (12% with less than 1% of anthropic land use, 49% with less than 15%). A maximum of 28 contaminants were found in a single lake, with 98% of lakes having between 0 and 12 different compounds (**Figure 5**) and an overall median of 3 detected compounds. Detection frequencies reported here are similar to those in previous large-scale studies on the occurrence of TrOCs in surface waters in the United States and in Europe (Bradley et al., 2017; Focazio et al., 2008; Kolpin et al., 2002; Loos et al., 2009).

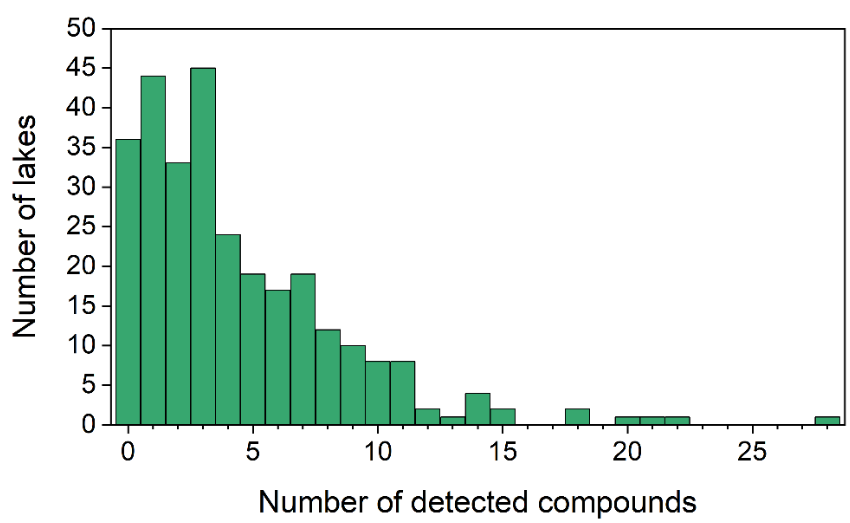

Figure 5. Distribution of the number of compounds detected per lake in the sampled Canadian lakes.

Pharmaceuticals were the most detected contaminants, but it is also the class including the highest number of targeted compounds (Annexe 1 **Figure 11**A and B). Out of the 27 tested pharmaceuticals, 25 were detected at least once, with an overall detection rate of 77%. Twenty of the detected pharmaceuticals were quantified with a median concentration of 9.1 ng/L. Additionally, this category accounted for 32% of the total contaminants' concentration (Annexe 1 **Figure 11**B). Among the most frequently detected sub-classes were stimulants, antibiotics, and analgesics.

Of the 16 pesticides that were targeted, 12 were quantified. Pesticides were also detected in more than half of the sampled lakes, with at least one of the 12 quantified pesticides found in 64% of lake water samples (Annexe 1 **Figure 11**A) with a median concentration of 15.6 ng/L. These included 9 herbicides and 3 insecticides and accounted for 25% of the total contaminant concentration (Annexe 1 **Figure 11**B). Except for the three targeted neonicotinoids, all of these compounds are among the most commonly used pesticides in agriculture in Canadian provinces (Murray et al., 2011).

Overall, the most detected compounds were the pesticide atrazine found in 45% of the sampled lakes, cotinine, the metabolite of the stimulant nicotine, with a 42% detection frequency, and deethylatrazine, the atrazine transformation product, quantified in 35% of sampled lakes. The other contaminants identified in more than 25% of the samples were the caffeine metabolite 1,7-dimethyxanthine (32%), the herbicide 2,4-D (31%), and the stimulant caffeine (27%).

In addition to the transformation products above, desmethylvenlafaxine, a metabolite of the antidepressant venlafaxine, was found at a frequency of 5.9% at concentrations up to 165 ± 30 ng/L while its parent compound venlafaxine was only detected in 2.8% of the lakes at up to 97.5 ± 1.4 ng/L. This trend has also been observed in U.S. surface and groundwaters with high metabolites detection frequencies (Bai et al., 2018; Focazio et al., 2008; Kolpin et al., 2002). These results highlight the importance of monitoring contaminants' metabolites in addition to parent compounds. Moreover, there is a lack of ecotoxicity data for these compounds, which could potentially be more harmful than their parent compounds. This has been demonstrated for other pharmaceuticals such as diclofenac (Diniz et al., 2015), cyclophosphamide (Russo et al., 2018), ribavirin (Wu et al., 2022), ibuprofen and naproxen (Grabarczyk et al., 2020).

Nevertheless, detection frequencies can be influenced by the method's limit of detection (LOD) (Fairbairn et al., 2016). No significant linear correlation was observed between the mean LOD and detection frequency in the present study (Annexe 1 **Figure 12**). Also, Welch's t-test showed that detection frequencies for compounds with LOD < 11 ng/L (the median value) and LOD > 11 ng/L were not significantly different ($p = 0.1140$). For some compounds, the method's limit of quantification (LOQ) seemed to have a major impact on the absence of quantification. This is the case for naproxen, chlorpyrifos, diazinon and triallate that had some of the highest mean LOQ (452, 317, 258 and 408 ng/L, respectively). These LOQ are above the median concentrations found in U.S. streams of 60 and 70 ng/L for chlorpyrifos and diazinon (Kolpin et al., 2002).

2.7.2. Concentration levels of TrOCs

The concentration of compounds typically ranged over 3 orders of magnitude for individual compounds and over 5 orders of magnitude overall (**Figure 6**). The quantification of each contaminant per lake is given in Annexe 1 **Tableau 12**. Overall, concentrations of quantified contaminants ranged from 0.23 ± 0.01 ng/L for cotinine in Dezadeash Lake, YT, to 2.2 µg/L (extrapolated to 4.5 ± 0.7 µg/L) for salicylic acid in Fenerty Lake, NS. This extrapolated concentration is close to maximum salicylic acid concentrations found in a recent occurrence assessment of a few Canadian surface waters at 4.4 µg/L (Pulicharla et al., 2021), as well as in the natural waters of other countries (Nannou et al., 2015; Peng et al., 2014). It is also in the same range as the median concentrations in WWTPs effluents in Canada, even though the maximum concentration in effluents were as high as 59.6 µg/L (Metcalfe et al., 2003). These high values could thus indicate the presence of close contamination sources such as wastewater discharges near that lake. The highest extrapolated concentration was also found in Fenerty Lake, NS, for sucralose at 18.1 ± 2.6 µg/L. Artificial sweeteners were the compounds with the highest detected concentrations in Great Lakes urban watershed's surface waters (Baker et al., 2022) and in Swedish lakes and rivers (Malnes et al., 2022) as well, with sucralose concentrations up to 630 and 370 ng/L respectively. The highest extrapolated sucralose concentration quantified in the present study (18.1 ± 2.6 µg/L) is in the same order of magnitude as concentrations found in a WWTP effluent at 12 µg/L by Baker *et al.* (Baker et al., 2022). This high quantification might be due to the presence of a WWTP in its watershed or to leaky individual septic systems of lakeshore habitations (James et al., 2016).

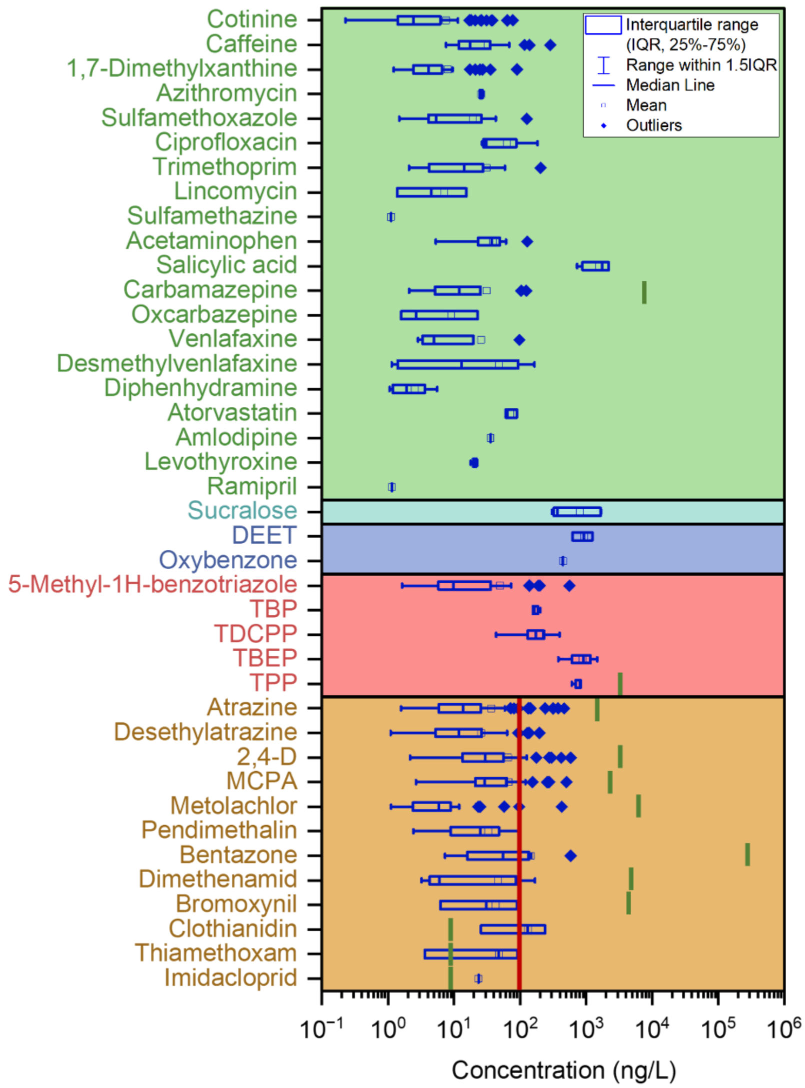

Figure 6. Concentrations measured in all lake samples for each contaminant. Boxplots represent quantified concentrations. Green text and background are used for pharmaceuticals, light blue for artificial sweeteners, blue for personal care products, red for consumer product additives, and orange for pesticides. The vertical lines represent thresholds according to the European drinking water directive (in red) (Council of the European Union, 1998) and from the Quebec province freshwater guidelines (in green) (MELCC, 2021). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Twenty-eight TrOCs (52% of targeted compounds) had quantification medians greater than 10 ng/L, 9 contaminants (17%) had medians over 100 ng/L, and salicylic acid had a median over 1 µg/L. The overall median was of 12 ng/L. Taken as a whole, about 58% of all quantified concentrations exceeded 10 ng/L, 15% were above 100 ng/L, and 3% above 1 µg/L. Moreover, cumulative concentrations of individual samples ranged from 0.23 ng/L to 8.2 µg/L with a median of 47 ng/L for lakes with at least 1 detection.

Contaminants were detected in the same concentration range in the survey of U.S. streams and medians of detected concentrations above 100 ng/L and 10 ng/L were found in proportions similar to the present study (11% and 39% respectively), even though more than 700 compounds were included in the survey (Bradley et al., 2017). However, they quantified 6 compounds with medians above 1 µg/L, while only salicylic acid exceeded that threshold in the present study. Moreover, the extrapolated maximum concentration in the samples collected in the present study (18.1 µg/L) is between the maxima found in U.S. surface and groundwaters at 2.4 µg/L for tetrachloroethylene (Focazio et al., 2008) and at more than 80 µg/L for 3,4-dichloroaniline (Bradley et al., 2017). However, the highest individual and cumulative concentrations in the sampled lakes (18.1 and 8.2 µg/L, respectively) are well below maxima found in a worldwide river contamination assessment, with concentrations up to 227 µg/L for acetaminophen and a cumulative pharmaceutical concentration of 297 µg/L in a highly impacted Bolivian river (Wilkinson et al., 2022).

In terms of comparing quantified values to existing thresholds, 8 of the 12 quantified pesticides were found at concentrations over the European guideline (Council of the European Union, 1998) of 100 ng/L for single pesticides in drinking water (**Figure 6**). This limit was exceeded in 7.2% of sampled lakes. Additionally, the 500 ng/L threshold for the summed pesticides' concentrations (Council of the European Union, 1998) was surpassed in 3.1% of lakes. The Quebec criteria for the protection of aquatic life in freshwater regarding chronical effects (MELCC, 2021), which have different values for each contaminant (Annexe 1 **Tableau 13**), was exceed for all 3 detected insecticides in 0.5 to 1.1% of lakes but it was not exceeded by any herbicide or pharmaceutical. However, the thresholds for herbicides and pharmaceuticals range from 1.8 to 510 µg/L, except for triallate which is at 240 ng/L, whereas the criteria for insecticides are below 10 ng/L.

Moreover, thresholds for the insecticides chlorpyrifos, diazinon, and neonicotinoids (2, 4, and 8.3 ng/L respectively) were lower than their mean LOQ that ranged from 12.4 to 96.1 ng/L. These criteria might be exceeded in lakes with concentrations below LOQ, leading to an underestimation of the extent of the potential impacts on aquatic organisms. Most pesticides targeted in this study exceeded at least one of the established guidelines and environmental thresholds in some lakes, suggesting that potential impacts on these lakes' ecosystems might occur.

2.7.3. Environmental risk assessment of TrOCs

2.7.3.1. Risk quotients for lake ecosystems

Even though target contaminants were usually detected at low concentrations, there could be non-negligible impacts on the lakes' ecosystems. For example, in the Great Lakes tributaries, with most TrOC median concentrations below 100 ng/L, Elliott *et al.* indicated elevated hazard predictions for 89% of studied river basins, most frequently due to the concentrations of the pharmaceuticals carbamazepine and venlafaxine, the fragrance galaxolide, and the phytosterol b-sitosterol (Elliott et al., 2021).

In the present study, risk quotients (*rq*) were calculated for single compounds using equation 1 and varied between 0 and 49. Maximum *rq* per compound are detailed in Annexe 1 **Tableau 14** and the detailed *rq* results per compound are given in Annexe 1 **Tableau 15**, **Tableau 16**, and **Tableau 17** for fish, daphnids, and ciliates, respectively. The lipid-lowering drug atorvastatin was found to pose a very high risk ($rq \geq 10$) to fish (*P. promelas*) with a maximum value of 45. Similarly, the flame retardants TDCPP and TBEP were found to pose a very high risk to daphnids (*D. magna*) with maximum values of 49 and 15, respectively. These results are in the same range as recently published risk assessments studies. An evaluation of the impact of pharmaceuticals on the world's rivers found high individual hazard quotients with values up to 28 for sulfamethoxazole in the Democratic Republic of Congo (Bouzas-Monroy et al., 2022). A study on Brazilian surface waters also found acute $rq \geq 10$ for several compounds (i.e., 4-n-nonylphenol, diclofenac and triclosan) (de Rezende and Mounteer, 2023). Moreover, both fish and daphnids exhibited high risk ($rq \geq 1$) in 10 lakes each for 3 different TrOCs. Overall, these results emphasize the importance of assessing the potential impacts of detected contamination, as even "low" concentrations seem to pose non-negligible risk to aquatic species.

Moreover, the detection of contaminant mixtures found in the sampled lakes suggests that risks of adverse effects on lakes' ecosystems could be underestimated by single compound risk assessment, as cocktail effects have been highlighted in several studies (Cedergreen, 2014; Gregorio and Chèvre, 2014; Hamid et al., 2021; Neale et al., 2020; Quinn et al., 2009). Thus, the summed risk quotient (RQ) of mixtures was estimated by adding the *rq* of all contaminants quantified in the sampled lakes. Of the 290 lakes analyzed, 17 (i.e., 6%) exhibited high potential risk ($RQ \geq 1$) for at least one of the aquatic species studied (**Tableau**

1). Four of those lakes (Kalamalka Lake, BC, Fenerty Lake, NS, Moore Lake, NS, and Falcon Lake, MB) showed high potential risk for both fish and daphnids. The highest RQ was found for *D. magna* in Kalamalka Lake, BC, with a value of 61, exhibiting a very high potential risk for this species. Ginebreda et al. also found the highest summed hazard quotient for *D. magna* at a similar level at 46 for the assessment of 29 pharmaceutical compounds in a Mediterranean river in Spain (Ginebreda et al., 2010). No RQ ≥ 1 was calculated for *T. pyriformis*, however, there was a lack of experimental and estimated toxicity values for this species, including for contaminants with high detection frequencies or concentrations, leading to an underestimation of the risk.

Tableau 1. Summed risk quotient (RQ) of lakes with a value of 1 or above for at least one of the three model species. RQ values ≥ 1 are presented in bold.

Lake	*P. promelas* (RQ)	*D. magna* (RQ)	*T. pyriformis* (RQ)
Kalamalka Lake, BC	**2.69**	**70.81**	0.21
Fenerty Lake, NS	**3.30**	**50.10**	0.30
Moore Lake, NS	**2.55**	**46.27**	0.29
Cedar Lake, ON	**45.00**	0.15	0.00
Falcon Lake, MB	**1.29**	**34.19**	0.14
Lac Brome, QC	**31.85**	0.11	0.00
Lac Wasi, ON	**30.34**	0.10	0.00
NA, SK	0.16	**12.49**	0.02
Little Lake, ON	0.11	**11.86**	0.00
Lac Innocent, NS	0.07	**7.91**	0.00
Stevenson Lake, ON	**7.53**	0.02	0.00
Lac des Indiens, QC	**7.18**	0.00	0.00
Lac Désiré, QC	**7.15**	0.00	0.00
Lac Winsch, QC	0.05	**6.46**	0.00
Pike Lake, ON	**6.00**	0.01	0.00
Boag Lake, AB	0.19	**4.12**	0.00
First Lake, NS	0.04	**3.99**	0.00

The estimated risks were driven by different contaminant classes depending on the species and some specific compounds appeared to be the main drivers of the estimated RQs (**Figure 7**). For fish, the risk was mostly linked to pharmaceuticals, more specifically to atorvastatin, levothyroxine, and ciprofloxacin. The flame retardants TPP, TDCPP, and TBEP, and the herbicide pendimethalin also impacted RQs for this species. For crustaceans, the risk was almost exclusively due to TDCPP, TBEP, and TPP, while ciliates were at moderate risk ($0.1 \leq RQ < 1$) from exposure to TPP and pendimethalin. The sensitivity to contaminants of each test species also varied in other risk assessment studies, with different pharmaceuticals being highlighted as the main contributors for fish, crustacean, and algae in Spanish rivers (Ginebreda et al., 2010). Thus, the drivers of estimated risks are highly species-dependent and are linked to the modes of action of the various contaminants. This stresses the importance of assessing risk for several species along the trophic levels to better estimate the global impact of TrOCs on ecosystems.

The use of an additive model to screen potential risk for lake ecosystems overlooks potential synergetic or antagonistic interactions between detected contaminants, which might lead to an under or overestimation of the actual risk (Rodea-Palomares et al., 2015). However, it represents an interesting approach to screen for lakes where contamination by TrOCs could be problematic and that require further ecotoxicological studies. Another limitation to this study is the use of estimated toxicity values as it adds to the uncertainty of the risk assessment results. In the present study, 44% of the values used were experimental. While less accurate than experimental values, estimated toxicity values obtained by QSAR methodologies remain necessary for a more comprehensive evaluation of potential impacts of contaminants in the environment (Schultz et al., 2003). In this study, the lack of estimated values for some contaminants in the case of *T. pyriformis* led to an incomplete risk analysis. It is thus necessary to expand experimental toxicity data for less studied species to improve predictive tools and allow for more extensive environmental risk assessments.

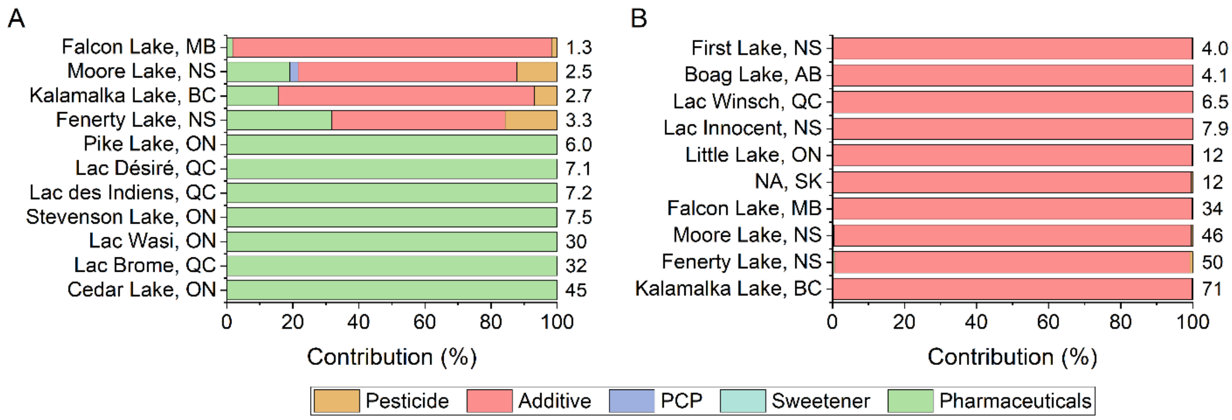

Figure 7. Percent contribution of contaminants' classes to RQ ≥ 1 for (A) the fish species *P. promelas* and (B) the crustacean *D. magna*. The RQ value is provided at the right of each bar.

2.7.3.2. Sub-lethal impacts of detected TrOCs

Sub-lethal impacts can also be expected as the detected TrOCs can be persistent and could thus induce chronic effects on aquatic biota. Potential sub-lethal effects were also evaluated using T.E.S.T. based on contaminants' detections. Estimations show that 37 of the 49 detected contaminants are potential developmental toxicants, and mutagenicity could be expected from 9 detected compounds (see Annexe 1 **Tableau 18**). Based on the detection of TrOCs in lake water samples, 59% of lakes could experience these sub-lethal effects, with up to 17 compounds with potential impact detected in a single lake. Even lakes with low calculated RQs could experience detrimental alterations in their ecosystems due to the presence of such contaminants. Although uncertain, these effects should not be neglected.

Moreover, the presence of antibiotics in the environment is of concern given their impact on aquatic microbiota. The detected contamination could lead to a selection of antibiotic-resistant microorganisms in some lakes as concentrations were found above PNECs, especially for ciprofloxacin that had a maximum concentration (i.e., 183 ± 19 ng/L) above its PNEC for resistance selection (i.e., 64 ng/L) (Bengtsson-Palme and Larsson, 2016). Moreover, the diversity of antibiotic resistance genes in Canadian lakes was demonstrated to be influenced by the use of antibiotics in their watersheds (Kraemer et al., 2022).

Regarding the high detection levels of sucralose, several studies have highlighted its absence of toxicity at concentration levels as high as 1 g/L (Huggett and Stoddard, 2011; Stolte et al., 2013). However, sub-lethal effects have been demonstrated at environmental concentrations on various aquatic species, such as changes in locomotion and respiration of gammarids and in the swimming height of daphnids exposed to 5 µg/L of sucralose (Wiklund et al., 2012), as well as genotoxic damage on carps at concentrations as low as 50 ng/L (Heredia-García et al., 2019). The present study found that these thresholds were exceeded in 2 and 9 lakes, respectively. Moreover, concomitant exposure to sucralose and caffeine can significantly increase anxiety-like behavior in fish hatchlings (Lee and Wang, 2015).

Previous studies have shown that concentrations of the UV filter oxybenzone as low as 100 ng/L can cause DNA damage in guppy fish (*Poecilia reticulata)* (Almeida et al., 2019), while concentrations as low as 22.8 ng/L can inhibit the growth of algae (*Chlorella* sp. and *Arthrospira* sp.) (Zhong et al., 2019). This indicates the potential harm that oxybenzone can cause to aquatic microorganisms in Moore Lake, NS, where it was measured at a concentration of 447 ng/L.

Disruption of ecosystem functioning is therefore expected for some sampled lakes as sub-lethal effects could occur at the concentration levels identified in this study.

2.8. Conclusions

This is the first extensive study on the occurrence and potential risks of TrOCs in lakes across Canada. The presence and concentration levels of 54 contaminants were investigated in 290 lakes, showing the detection of at least one TrOC in 88% of investigated lakes, with the presence of mixtures of compounds in most lakes. Atrazine, deethylatrazine and cotinine were the most detected compounds, found in more than a third of the lakes. However, the highest concentrations were found for sucralose at contamination levels approaching those found in WWTP effluents, while most targeted pesticides exceeded at least one of the established guidelines and environmental thresholds in some lakes, suggesting that potential impacts on these lakes' ecosystems might occur. In fact, even though most TrOCs were quantified at low concentration levels, the environmental risk assessment highlighted high potential risk from mixtures of contaminants in 6% of the studied lakes, with various TrOCs driving the risks depending on the target species. Sub-lethal effects are also expected for some compounds in up to 59% of lakes.

This study aimed to fill knowledge gaps about the presence of TrOCs in Canadian lakes. It provides a baseline to monitor the evolution of contamination in these water bodies. Furthermore, the data obtained can help improve policies for the protection of aquatic ecosystems. Additionally, the results can be employed by local authorities to make informed decisions on prioritizing lakes for further investigation of contamination sources and conservation efforts. In a follow-up study, the distribution of TrOCs across Canada and the main factors influencing lake contamination will be addressed.

2.9. Author information

2.9.1. Notes

The authors declare no competing financial interest.

2.10. CRediT authorship contribution statement

Lisa Lahens: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing - Original Draft, Writing - Review & Editing, Visualization. **Hubert Cabana**: Resources, Writing - Review & Editing. **Yannick Huot**: Conceptualization, Methodology, Resources, Writing - Review & Editing, Funding acquisition. **Pedro A. Segura**: Conceptualization, Methodology, Resources, Writing - Review & Editing, Supervision.

2.11. Declaration of competing interest

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

2.12. Data availability

Data will be made available on request.

2.13. Acknowledgments

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2.14. Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123764 (Annexe 1).

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CHAPITRE 3. INFLUENCE DES ACTIVITÉS ANTHROPIQUES SUR LA CONTAMINATION ORGANIQUE TRACE DES LACS CANADIENS

3.1. Notes préliminaires

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3.3. Contribution des auteurs

L.L. et P.A.S. ont conçu le projet. L.L. a élaboré les hypothèses de travail et a effectué les analyses statistiques préliminaires. L.L. et J.A.C. ont développé et validé les modèles de régression. L.L. a effectué l'interprétation des résultats et a rédigé le manuscrit. L.L., J.A.C., H.C., Y.H. et P.A.S. ont révisé le manuscrit. Tous les auteurs ont contribué à cet article et ont approuvé la version finale.

3.4. Abstract

Anthropogenic activities and urbanization often lead to the discharge of organic compounds into surface waters. Previous exploratory studies have brought forward potential drivers of this contamination. Yet, to better mitigate the contamination of natural waters and prioritize protection efforts, it is imperative to investigate these relationships further. This study aimed to verify the effect of specific anthropogenic factors on water contamination caused by trace organic contaminants (TrOCs) such as pharmaceuticals and pesticides. Data on the detection and concentration levels of TrOCs, major anthropogenic land use, and human activities from a large-scale study on Canadian lakes were used to reach this goal. The impact of agricultural and urban land use, the presence of wastewater treatment plants and hospitals in lakes' watersheds, and population and livestock densities on lake water contamination was investigated by applying negative binomial and ordinal logistic regression models. The analysis confirmed that agriculture, urbanization, and the presence of wastewater treatment plants in lake watersheds have a predominant influence on their contamination. The effects of these factors varied depending on the contaminant classes. Moreover, unsampled Canadian lakes with high anthropogenic pressure were brought to light as potential candidates for monitoring studies as their waters could be at risk of organic contamination. Overall, these results demonstrate that even in complex ecosystems such as lakes, it is possible to use a limited number of factors to explain anthropogenic contamination. This can help policymakers make informed decisions on contamination mitigation and provide insights into watershed management.

Keywords: Human impact; land use; PPCPs; pesticides; consumer product additives

3.5. Introduction

A wide variety of chemicals are used daily to tackle various issues. Often, these compounds find their way into the environment, accumulating more specifically in aquatic ecosystems (Kolpin et al., 2002). Lakes, for instance, are known to integrate compounds that are released in their watersheds due to surface runoff (Müller et al., 1998) or direct discharge in tributaries. The type of compound entering these ecosystems thus depends on the anthropogenic activities taking place in their watersheds. Trace organic contaminants (TrOCs) finding their way to surface waters include pesticides, pharmaceutically active compounds, food additives, personal care products, and industrial additives such as flame retardants and plasticizers (Gerbersdorf et al., 2015; Sauvé and Desrosiers, 2014).

The presence of TrOCs in surface waters can induce adverse effects on ecosystems and human health. Indeed, these contaminants are usually persistent or pseudo-persistent, as they are continuously discharged into the environment, are highly mobile and can exert toxicity in the environment (Montes et al., 2022; Richmond et al., 2017). TrOCs, in particular pharmaceuticals and personal care products (PPCPs), are known to disturb environmental ecosystems due to effects such inhibition of organisms' growth and alteration of community composition (Richmond et al., 2017). Environmentally relevant concentrations have been found to impact aquatic organisms such as algae, crustaceans, and fish (Gergs et al., 2013; Kidd et al., 2007; Malaj et al., 2014).

The presence and concentrations of contaminants are unevenly distributed among lakes. TrOCs have been found in lakes around the world at concentrations from low ng/L to several µg/L (Katsikaros and Chrysikopoulos, 2021). Concentrations are usually higher in Asia (Chen and Ma, 2021), while a greater number of contaminants are often found in Europe and North America (Anagnostopoulpou et al., 2022; Focazio et al., 2008). However, dissimilarities within the same country are also observed, in part because the entry of contaminants into surface waters is controlled by watershed characteristics such as the presence of important point sources (Scown et al., 2017; Segura et al., 2015; Wilkinson et al., 2024). Moreover, lakes and their watersheds sustain a high complexity of processes involved in contaminants transport and natural attenuation such as surface runoff, leaching, hydrolysis, photolysis, biodegradation and association with dissolved organic matter (Berkowitz et al., 2014; Blunt et al., 2018; Hung et al., 2006; Kim et al., 2014). It is thus necessary to understand the drivers of contamination distribution to improve watershed management.

Previous studies have shown a correlation between potential environmental and anthropogenic factors and contaminants' detection and concentrations. Land use has been identified in multiple studies as a significant factor affecting the frequency of detection and concentration levels of contaminants. Watersheds impacted by agricultural or urban uses show higher levels of detection and concentrations of contaminants (Baldwin et al., 2016). Both human and livestock density are also potential drivers of contamination, with population density impacting the detection and concentrations of most TrOCs classes and livestock density influencing antibiotic levels (Baldwin et al., 2016; Johnson and Bell, 2023; Servadio et al., 2021). Point sources such as wastewater treatment plants (WWTPs), septic systems, or hospitals have also been noted as important contamination sources (Servadio et al., 2021; Verlicchi et al., 2012). Even though most research focuses on streams, where sources are often easily identifiable, the identified correlations can be tested for lakes as similar watershed characteristics might influence both surface water types.

There is a need to bring to light the main drivers of lake water contamination despite the multitude of factors involved and the high complexity of these ecosystems. The main goal of this study was thus to verify the association of specific factors with the occurrence and concentrations of 54 targeted TrOCs including pesticides, PPCPs and consumer product additives, and to quantify the significance of their effects using statistical inference. To meet this goal, we used data on TrOCs and anthropogenic factors from a nationwide study of Canadian lakes (Huot et al., 2019; Lahens et al., 2024) to answer specific hypotheses developed based on suspected contamination drivers:

Hypothesis #1: The number of TrOCs detected in lake water and their total summed concentrations will increase with the fractions of urban and agricultural land use, the population density, and the presence of WWTPs in lakes' watersheds.

Hypothesis #2: The summed concentration of pesticides in lake water will increase with the fractions of urban and agricultural land use, and the population density in lakes' watersheds.

Hypothesis #3: The summed concentration of pharmaceuticals, personal care products, and consumer products additives in lake water will increase with the fractions of urban land use, the population density, and the presence of hospitals and WWTPs in lakes' watersheds.

Hypothesis #4: The summed concentration of antibiotics in lake water will increase with the population and livestock densities, and the presence of hospitals and WWTPs in lakes' watersheds.

3.6. Material and methods

3.6.1. Data sources

The data used in the present analysis come from the Lake Pulse survey, a large-scale Canadian study on lake health. The collection of all variables is described in the Lake Pulse field manual (LakePulse, 2021). Briefly, lake selection was done with a stratified random selection based on ecozone, lake size (small from 0.1 to 0.5 km^2, medium from 0.5 to 5 km^2, and large from 5 to 100 km^2), and a human impact index calculated from land use data on lakes' watersheds to allow for the analysis of lakes representative of distribution in Canada. Watersheds were divided into pixels, each assigned a human impact value depending on land-use activity. Natural landscapes had a value of 0, forestry and pasture were assigned 0.5, and urbanization, agriculture, and mining were attributed a value of 1. The human impact index was then calculated as the average of land use pixels forming the lake's watershed, ranging from 0 to 1 and lakes were separated by impact categories (low, moderate, and high). The cutoff values between categories vary between ecozones as they were set to have an equal number of lakes in each category for each ecozone. Details on lake selection have been described previously (Huot et al., 2019).

For the present study, only lakes where TrOCs concentrations were assessed in the surface layer (epilimnion) are considered (**Figure 8**). Details on these lakes are given in Annexe 2 **Tableau 19**. A summary of data acquisition and of the main characteristics of the variables considered in the present study are given below, and their values are given in Annexe 2 **Tableau 20**.

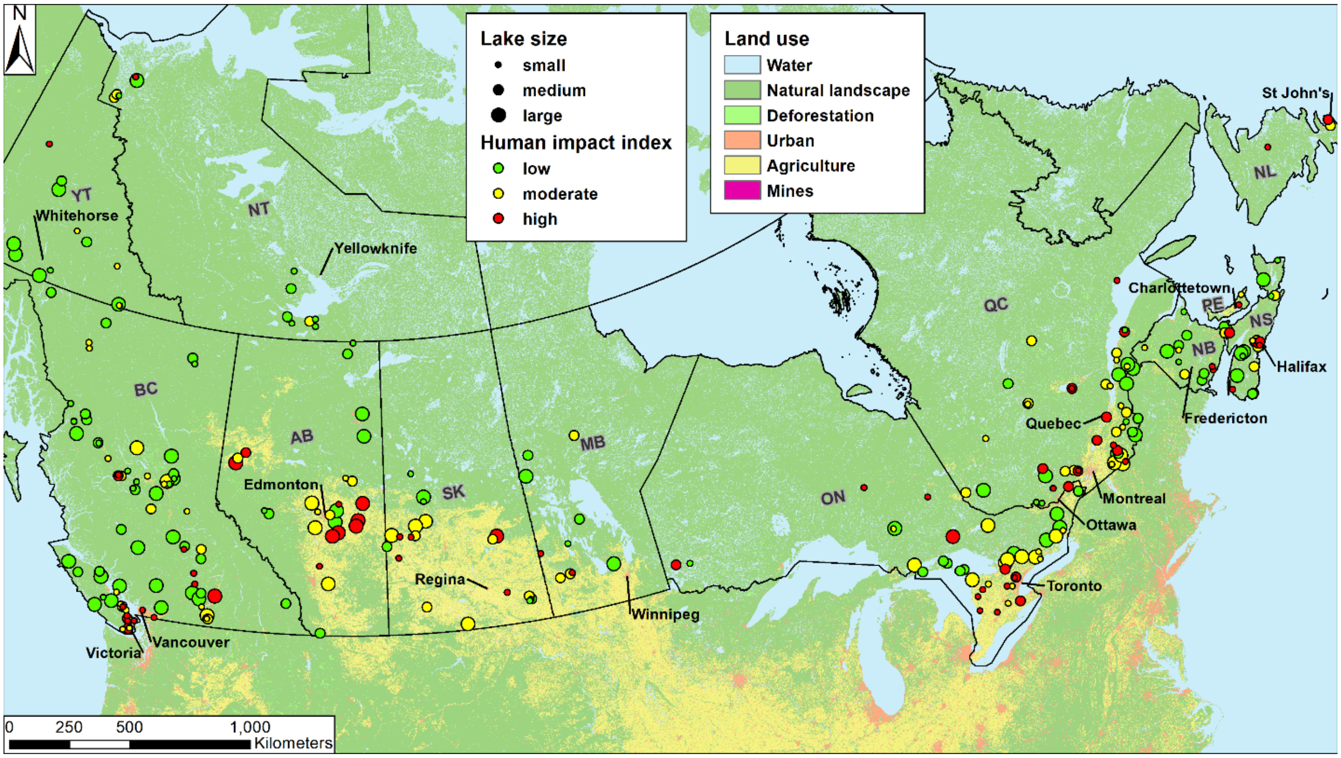

Figure 8. Location of the 290 sampled lakes. Lake size and human impact index are represented as circle size and color, respectively. The map's background represents the land use type. Canadian provinces and territories: YT: Yukon, NT: Northwest Territories, BC: British Columbia, AB: Alberta, SK: Saskatchewan, MB: Manitoba, ON: Ontario, QC: Quebec, NB: New Brunswick, PE: Prince Edward Island, NS: Nova Scotia, NL: Newfoundland and Labrador.

3.6.2. Dependent variables

The response variables are composed of the results from the analysis of 54 contaminants representative of various human activities such as agriculture, farming, urban development, industry, and recreational activities. The studied TrOCs were chosen for their ubiquitous presence in the environment (Bradley et al., 2017; Focazio et al., 2008; Kolpin et al., 2002), their toxicity (Ortiz de García et al., 2013), and their extensive use (Murray et al., 2011). The methods and results have been described previously (Lahens et al., 2024). Briefly, sub-surface samples were collected in amber high-density polyethylene bottles between June and September in 2017, 2018 and 2019. Samples were rapidly stored at -20°C to limit degradation. In the laboratory, TrOCs were analyzed in triplicates by liquid chromatography-triple quadrupole mass spectrometry (LC-MS/MS) after sample preconcentration. Lake water samples were filtered and pH-adjusted to 6.5 before concentration by solid phase extraction at a concentration factor of

500. Extracts were spiked with internal standards and evaporated to dryness before reconstitution in H_2O-MeOH (80:20, v/v). The samples were then analyzed by LC-MS/MS in positive and negative ionization modes. The targeted compounds were pesticides (10 herbicides and 6 insecticides), pharmaceuticals and personal care products (PPCPs) (27 pharmaceutically active compounds, 3 personal care products) as well as consumer product additives (1 food additive, 7 industrial additives). Antibiotics included in the analysis were for human (3), veterinary (3), or mixed (5) use. Quality control procedures, such as sample and laboratory blanks, limits of detection and quantification, calibration controls and relative standard deviation thresholds, were applied to ensure the reliability of the obtained data. The final concentrations were corrected by extraction recoveries to give a better estimate of the true environmental concentration.

To summarize the results previously published, 40 compounds out of the 54 targeted were detected at least once in 88% of the 290 sampled lakes and quantified in 68% of them (Lahens et al., 2024). The number of detections and the summed concentration of all TrOCs, as well as the sums of pesticides, PPCPs and consumer product additives, and antibiotics, were selected as response variables in the present study. Across all lakes, the detected number of compounds ranged from 0 to 28, with combined concentrations of measured TrOCs between 0 to 8162 ng/L. The sum of all pesticide concentrations was calculated and values up to 1386 ng/L were found in one lake. For PPCPs and additives, their sum reached up to 7636 ng/L. Antibiotics were also identified, with their sum reaching up to 513 ng/L.

3.6.3. Independent variables and control variables

The descriptive statistics for the lake characteristics relevant to this study are presented in **Tableau 2**. Several independent variables were selected according to a review of the literature and data availability. Land use fractions were determined as the fractions of lakes' watersheds assigned to agriculture and urban development. Population density in the lakes' watersheds was calculated based on population data for the respective sampling years. The presence of WWTPs and hospitals in the watersheds were included as categorical variables. Lastly, livestock density was computed based on animal count data from 2016.

Control variables were also included in the statistical models. This designates variables that are not of interest to address the present hypotheses. However, they must be included in the models as they might affect the results for the outcomes of interest. The area ratio between a lake and its watershed, and lake depth were included as they can impact contaminants' dilution. The area ratio was calculated as the lake area divided by its watershed area. Maximum lake depth was measured in the field using a depth finder. In addition, residence time was included as it might also affect the results as higher water retention can lead to contaminant accumulation. Average residence times were retrieved from the HydroLAKES database (Messager et al., 2016). For six lakes, a residence time could not be calculated because of the absence of discharge. Therefore, a value of 45500 days, corresponding to the highest calculated residence time in the dataset rounded up to the hundred, was assigned to those lakes. Control variables also included watershed slope and precipitation, which can lead to increased transport of TrOCs through runoff. The mean slope on the watershed was calculated as the mean of the percent rise inside the watershed. Precipitation was calculated as the sum of large-scale precipitation and convective precipitation over the seven days before sampling. Finally, the sampling date was added as day of the year as some compounds (e.g., agricultural pesticides) have varied application dates.

Tableau 2. Statistics of relevant lake characteristics.

Lake characteristics	Mean (SD) or n (%) *	Range
Agricultural fraction	0.08 (0.18)	0 – 0.88
Urban fraction	0.08 (0.15)	0 – 0.83
Population density (u/km²)	70 (313)	0 – 3704
Presence of WWTPs	20 (7%)	NA
Presence of hospitals	16 (6%)	NA
Livestock density (people/km²)	5 (9)	0 – 53
Area ratio	0.10 (0.10)	0 – 0.60
Lake depth (m)	14 (17)	0.65 – 138
Residence time (days)	2877 (8053)	0.10 – 45500
Mean slope (%)	8 (10)	0.07 – 62
Precipitation (cm)	2 (2)	0 – 8
Sampling date	NA	June 29 – September 12

* SD: standard deviation; n: number of occurrences for presence data.

3.6.4. Statistical models

A generalized linear model for count data was employed to investigate the effect of urban and agricultural fractions, the presence of WWTPs, and population density on the average number of detected TrOCs in lake water across Canada. Preliminary modelling of the data showed that the distribution of the number of detected TrOCs presented overdispersion, i.e., the variance of the distribution was considerably larger than its mean. Therefore, the generalized linear model assumed the number of detected TrOCs had a negative binomial distribution (Schober and Vetter, 2021). The regression model can be written as:

$$ln(\mu_i) = \beta_0 + \beta_1 x_{i1} + \beta_2 x_{i2} + \cdots + \beta_p x_{ip} \qquad [3.1]$$

where μ_i denotes the conditional mean number of detected TrOCs, also called incidence rate, for lake i, β_0 is the y-intercept, x_{i1}, x_{i2}, …, x_{ip} are the values of the independent and control variables for lake i, β_1, β_2, …, β_p are the coefficients to be estimated by the model, and *ln* denotes the natural logarithmic function. Results are interpreted in terms of incidence rate ratios (IRR) and 95% confidence intervals (95%CI). For categorical variables, the IRR compares the incidence rate for each category as compared to a reference category. For example, in the case of the categorical variable of WWTPs, the IRR compares the incidence rate for the presence of these structures as compared to their absence. For continuous variables, the IRR compares the incidence rate associated with a one-unit increase in the distribution of the variable. For instance, a 10% increase was employed for land use. The IRR also provides information on the size of the effect.

The model was adjusted for area ratio, lake depth, residence time, mean slope, precipitation, and sampling date. Lake depth, residence time, and mean slope were log-transformed to stabilize the variance or their distributions. Model fit, including checking for uniformity, overdispersion, zero-inflation, and the presence of possible outliers, was assessed by graphical inspection of randomized quantile residuals using a simulation-based approach (Dunn and Smyth, 1996). The R script and validation results are available in the Annexe 2 (section **A2.1** and **Figure 13**).

Ordinal logistic regression models were used to investigate the effect of the independent variables of interest, adjusting for the selected control variables, on the summed concentration of all TrOCs and of contaminants' classes. Although ordinal logistic regression was designed for categorical ordinal data (distributions where there is an intrinsic order), it also applies to continuous data, since they are also ordinal. The model can handle the non-detectable observations by modelling a mixture of a discrete and a continuous distribution and is thus applicable to measurements with detection limits, as the outcomes only need to be orderable. The usual approach of using logistic regression after binarizing the outcome as detectable and undetectable measurements ignores the information provided by the values of the distribution of detectable measurements. Furthermore, there is no need to replace non-detections with simulated values (Liu et al., 2017).

For the summed concentration of TrOCs, the independent variables of interest were urban and agricultural fractions, the presence of WWTPs, and population density. For the pesticides' sum, the independent variables included were urban and agricultural fractions, and population density. For the PPCPs and additives' sum, the independent variables were the urban land use fraction, the presence of hospitals and WWTPs, and population density. Finally, for the antibiotics' sum, the independent variables were livestock density, the presence of WWTPs, and population density. The presence of hospitals could not be included as no hospitals were present in watersheds of lakes with antibiotics quantifications.

Similarly to the negative binomial regression model, the ordinal logistic regression models were adjusted for area ratio, lake depth, residence time, mean slope, precipitation, and sampling date. Lake depth, residence time, and mean slope were log-transformed to stabilize their variance. A preliminary modelling of the data to compare log-likelihoods for models using the *probit*, *logit* and *loglog* functions showed that logit models provided the best fit. Further model fit and investigation of the presence of possible outliers were assessed by graphical inspection of probability-scaled residuals (PSR), including residual-by-predictor plots (Liu et al., 2017). The R script for the summed concentration of TrOCs and the validation results of all ordinal logistic regression models are available in Annexe 2 **A2.2**, **Figure 14**, **Figure 15**, **Figure 16**, and **Figure 17**.

In general, the ordinal logistic regression models use a logistic link function and can be written as:

$$ln\left(\frac{Pr(Y>y_i)}{Pr(Y\leq y_i)}\right) = \beta_{0i} + \beta_1 x_{i1} + \beta_2 x_{i2} + \cdots + \beta_p x_{ip} \quad [3.2]$$

where y_i is the value of the dependent variable Y (class or total sum of TrOCs) for lake i, β_{0i} is the y-intercept, x_{i1}, x_{i2}, …, x_{ip} are the values of the independent and control variables, β_1, β_2, …, β_p are the coefficients to be estimated by the model, and *ln* represents the natural logarithmic function.

The quantity $\frac{\Pr(Y>y_i)}{\Pr(Y\leq y_i)}$ represents the odds of having an exceedance probability as compared to a non-exceedance probability at the value $Y = y_i$. Results are interpreted in terms of odds ratio (OR), where an OR > 1 implies higher odds of having an exceedance probability as compared to a non-exceedance probability. For categorical variables, the OR compares the odds of exceedance for each category as compared to a reference category (e.g., the absence of WWTPs). For continuous variables, the OR compares the odds associated with a one-unit increase in the distribution of the variable (e.g., a 10%-increase in anthropogenic land use). The OR also provides information on the size of the effect.

Analyses were performed using the R software (v4.3.2; R Core Team, 2023). For the negative binominal regression, the *MASS* package (v7.3-60.2; Ripley, 2024) was employed. For the ordinal logistic regression, the *orm* function in the *rms* package (v6.7-1; Harrell Jr, 2023) was used. For the simulation-based quantile residual analyses of the negative binomial regression, the *DHARMa* package (v0.4.6; Hartig, 2022) was used.

3.7. Results and discussion

3.7.1. Distribution of contaminants on the Canadian territory

The TrOCs results have been previously described (Lahens et al., 2024). Here, we provide an overview of the relationship between contamination and anthropogenic land use. The distribution of TrOCs in Canadian lakes reveals more intense contamination in provinces covering the Canadian Prairies (i.e., Manitoba, Saskatchewan, and Alberta) and the Mixedwood Plains (i.e., Quebec and Ontario), where agriculture is more extensive, and urbanization is denser (**Figure 9**). Of the 12 lakes exhibiting total summed concentrations above 1 μg/L (**Figure 10**), eight are in those more human-impacted regions. Moreover, the highest number of detections and summed concentration were found for an urbanized lake (25% watershed with urban land use) near Nova Scotia's capital, Halifax. This lake was mostly contaminated by PPCPs and additives, like all lakes with a total concentration above 1 μg/L, except for 2 lakes in Ontario that were more impacted by pesticides. These 2 Ontarian lakes are located in watersheds with an important proportion of agricultural land (37 and 71%, respectively).

Statistical analysis was thus carried out to confirm and further investigate the impact of anthropogenic variables on the occurrence of TrOCs.

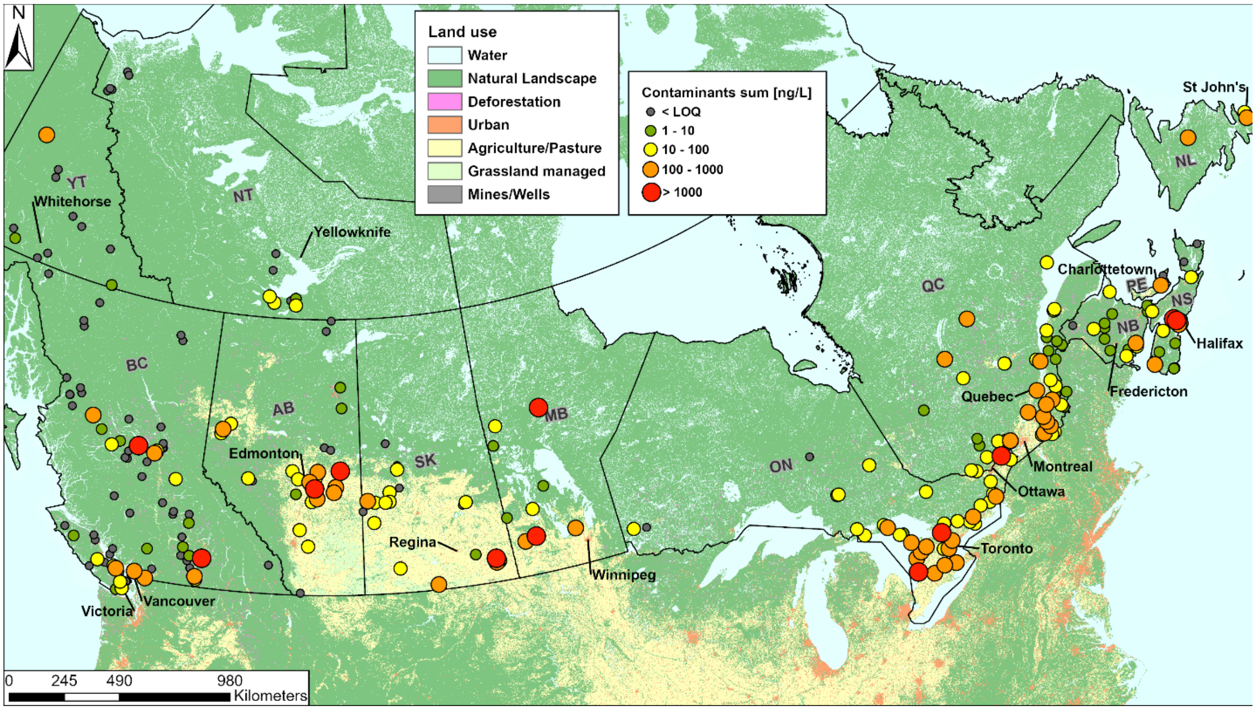

Figure 9. Distribution of the summed concentrations of all TrOCs in sampled lake waters across Canada (n = 290).

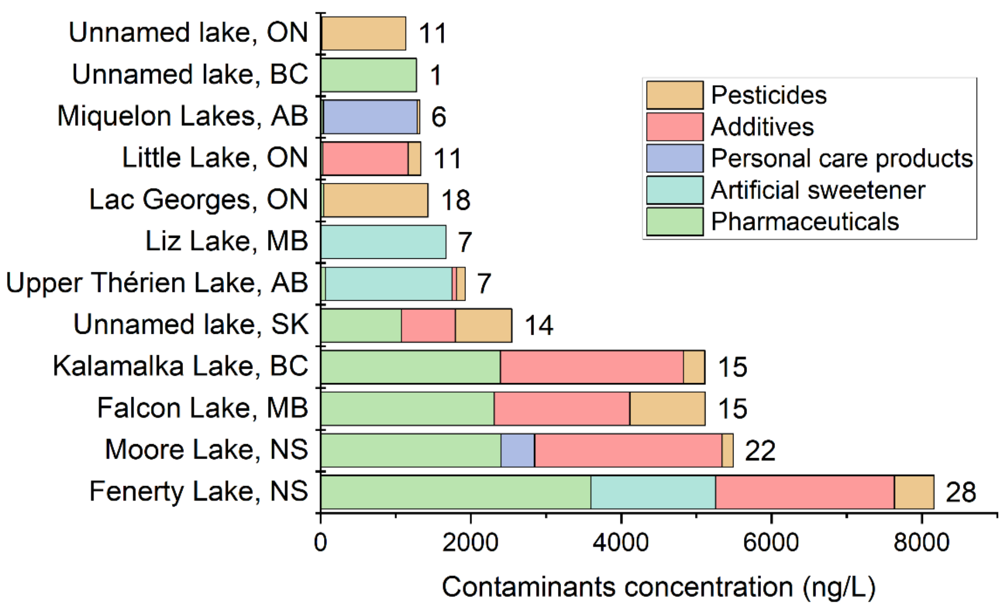

Figure 10. The stacked histograms represent the summed concentrations for each compound category for lakes displaying summed concentrations greater than 1 µg/L (red points in **Figure 9**).

3.7.2. Impact of human activities on contamination

On each of the regression models, model fit diagnostics and residual analyses showed that the variable population density presented high residual variability as well as high collinearity with the variable urban land use and was thus removed from the models. Therefore, its impact on the detection of the targeted TrOCs could not be assessed, but it is expected to associate with dependent variables in the same direction as urban land use.

3.7.2.1. Factors influencing the number of detections and total TrOCs concentration in lake waters

The results of the negative binomial regression model performed to evaluate the association of anthropogenic factors with the number of TrOCs detected in lakes are presented in **Tableau 3**. For this regression model, one lake (Moore Lake, NS) was highlighted as a possible outlier and was thus removed from the analysis. This lake could have other major sources of contamination than the ones included in the present analysis, such as forestry, recreational use, and possibly local businesses not following environmental regulations, or atmospheric depositions from nearby sources outside the watershed, thus leading to a higher number of compounds detected than expected.

Results showed that, in the presence of the other independent and control variables, the presence of WWTPs and the proportion of agricultural and urban land use in lakes' watersheds had a significant association with the number of TrOCs detections in the sampled lakes. The presence of WWTPs (IRR 2.72, $p < 0.001$) demonstrated a higher effect than land use percentages, with urban fraction having a slightly greater influence (10%-increase IRR 1.17, $p < 0.001$) than agriculture (10%-increase IRR 1.11, $p < 0.001$). The higher impact of WWTPs could be explained by the high concentrations discharged into the environment that might lead to higher detections, while the greater effect of urbanization over agricultural land use might be due to the larger number of urban contaminants rather than pesticides in the analysis, as 38 PPCPs and additives were included as compared to 16 pesticides. Additionally, several pesticides are used in urban settings while PPCPs and additives are seldom used on agricultural lands which can accentuate the higher effect of urban land use on TrOCs detection.

Tableau 3. Negative binomial regression results of covariates of interest on the number of detections ($n = 283$) *

Variable	β	Standard error	z-value	p-value **	IRR (95%CI) **
Agricultural fraction	0.01	0.003	3.86	**< 0.001**	**1.11** (1.05-1.18)
Urban fraction	0.02	0.003	5.06	**< 0.001**	**1.17** (1.10-1.25)
WWTPs presence	1.00	0.17	5.84	**< 0.001**	**2.72** (1.95-3.85)

*Model was adjusted for area ratio, lake depth, residence time, mean slope, precipitation, and sampling date (see outcomes in Annexe 2 **Tableau 21**). Six lakes had missing values of residence time.
**Values shown in bold represent a statistically significant association.

The results of the ordinal logistic regression model used to investigate the factors driving TrOCs summed concentrations are shown in **Tableau 4**. For these results, an OR > 1 implies higher odds of having an increased summed concentration than an equal or reduced concentration. Results showed that in the presence of the other independent and control variables, the higher the urban land use fraction in the lake's watershed, the higher the odds of exceedance of the summed concentration of TrOCs (10%-increase OR 1.58, $p < 0.001$). The agricultural land use fraction also had a similar impact (10%-increase OR 1.26, $p < 0.001$). Furthermore, the odds of exceedance of the summed concentration of TrOCs were higher in the presence of wastewater treatment plants (OR 6.95, $p < 0.001$). Regarding urbanization and agriculture, their impacts on the summed concentrations of TrOCs might be due to different reasons. Urbanization might introduce a higher number of contaminants whose concentrations add up, while agriculture could discharge fewer compounds in higher quantities.

The hypothesis on the impact of anthropogenic land use and activities in lakes' watersheds on their overall contamination (*vide supra* hypothesis #1) was tested using two separate models for the number of TrOCs detections and their summed concentrations The impact of urbanization, agriculture, and the presence of WWTPs was validated with the present dataset. Agricultural and urban land use, as well as the presence of WWTPs in the lakes' watersheds, influenced both the number of detections and summed concentrations of TrOCs. These outcomes support previous research that highlighted WWTPs (Servadio et al., 2021) and land use cover (Baldwin et al., 2016) as major estimators of detection and concentration levels of TrOCs in lakes and streams, respectively. Both studies also found population or population density in the watershed to be one of the main estimators for contamination. However, its effect could not be assessed in the present study. Nonetheless, the impact of population density may be included in the urban land use variable as these variables are highly correlated, and higher urbanization implies a greater population density in the watershed.

Tableau 4. Ordinal logistic regression results of covariates of interest on summed concentrations of total TrOCs and by class (n = 284) *

Variable	β	Standard error	z-value	p-value **	OR (95%CI) **
Total TrOCs sum					
Agricultural fraction	0.02	0.01	53.49	**< 0.001**	**1.26** (1.11, 1.44)
Urban fraction	0.05	0.01	5.99	**< 0.001**	**1.58** (1.36, 1.84)
WWTPs presence	1.94	0.41	4.74	**< 0.001**	**6.95** (3.12, 15.5)
Pesticides sum					
Agricultural fraction	0.05	0.01	6.04	**< 0.001**	**1.58** (1.36, 1.83)
Urban fraction	0.02	0.01	2.26	**0.02**	**1.19** (1.02, 1.39)
PPCPs and additives sum					
Urban fraction	0.05	0.01	6.13	**< 0.001**	**1.57** (1.36, 1.82)
WWTPs presence	1.72	0.54	3.17	**0.002**	**5.57** (1.92, 16.1)
Hospitals presence	0.14	0.64	0.22	0.8	1.15 (0.33, 4.00)
Antibiotics sum					
Livestock density	0.02	0.02	0.84	0.4	1.01 (0.98, 1.05)
WWTPs presence	0.66	0.63	1.06	0.3	1.94 (0.57, 6.65)
Population density	-0.0003	0.001	-0.41	0.7	1.00 (1.00, 1.00)

*Models were adjusted for area ratio, lake depth, residence time, mean slope, precipitation, and sampling date (see outcomes in Annexe 2 **Tableau 21**). Six lakes had missing values of residence time.
**Values shown in bold represent a statistically significant association.

3.7.2.2. Drivers of contaminants' classes concentration levels in lake water

The results of the ordinal logistic regression models applied to pesticides, PPCPs and additives, and antibiotics sums are shown in **Tableau 4**. Regarding the sum of pesticides, agricultural land use showed a highly significant effect (10%-increase OR 1.58, $p < 0.001$). Urban land use also affected pesticide levels (10%-increase OR 1.19, $p = 0.024$).

The hypothesis on the impact of agricultural and urban land use on lakes' pesticide contamination (*vide supra* hypothesis #2) was tested with an ordinal logistic regression model. The impact of agriculture and urbanization on pesticides sum was validated in Canadian lakes. The agricultural land cover, identified as a major predictor of detection and concentration of herbicides in U.S. streams (Baldwin et al., 2016), showed a highly significant effect on pesticide concentration levels in the present study. The potential importance of both urban and agricultural land covers on pesticides' concentration highlighted for streams in the Great Lakes area (Baldwin et al., 2016) was also confirmed by this study. The impact of urban land use can be explained by the frequent use of pesticides outside of agriculture (Kolpin et al., 2002). Indeed, pesticides used in urban settings, such as the herbicides 2,4-D and atrazine, and the insecticides chlorpyrifos and diazinon used on lawns, golf courses, and roads, were quantified in Canadian lakes. These pesticides can enter the environment directly through surface runoff without water treatment as opposed to other TrOCs such as pharmaceuticals. The effect of urban land use on pesticide concentrations suggests that contamination reduction efforts for these compounds should not only focus on agricultural applications but also consider non-agricultural uses, as they can significantly impact environmental loads.

Furthermore, the sum of PPCPs and additives was significantly associated with urban land use fraction (10%-increase OR 1.57, $p < 0.001$) and the presence of WWTPs (OR 5.57, $p < 0.001$) (**Tableau 4**).

An ordinal logistic regression model was also employed to test the hypothesis on the impact of urban land use and the presence of WWTPs and hospitals in lake watersheds on their PPCPs and additives concentrations (*vide supra* hypothesis #3). The impact of urbanization and WWTPs presence on PPCPs and additives lake contamination was validated with the available data, however, the impact of hospitals presence was not confirmed. These results agree with previously published studies. According to Baldwin et al. (2016), urban land cover is one of the main drivers of pharmaceuticals and additives concentrations in streams, while WWTP effluents are also major predictors for some types of TrOCs (e.g., flavours and fragrances). The same impact was observed for PPCPs and additives in Canadian lakes with higher concentrations expected with an increase in urban land cover and with the presence of WWTPs in lake watersheds. WWTPs have also been shown as important factors for pharmaceuticals' detection in an exploratory study on lake contamination (Servadio et al., 2021). These results stress the importance of reducing the contamination load of WWTPs effluents. Similar to the results obtained by Servadio et al. (2021) on the impact of healthcare facilities, the effect of the presence of hospitals was not significant.

However, reduction efforts could be enhanced by improving hospital effluents pre-treatment as they are known to largely contribute to WWTPs loads (Boucher et al., 2021; Lu et al., 2019; Verlicchi et al., 2012).

The hypothesis on the impact of WWTPs, livestock, and human presence on antibiotics concentrations (*vide supra* hypothesis #4) was also tested with an ordinal logistic regression model (**Tableau 4**). However, this hypothesis could not be validated with the present dataset. Indeed, no independent variable had a significant impact on the antibiotics sum. This can be due to the low frequency of quantification for this class of contaminants.

To our knowledge, this is the first study using ordinal logistic regression for a continuous outcome in the field of water contamination. Nevertheless, environmental sciences could benefit from implementing such models more frequently as they allow for the inclusion of data below detection limits in the assessment of driving factors. The high proportion of data below detection and quantification limits is a limitation of this study, for example in the case of antibiotics sum that had only 12% of quantified values. However, with 32, 53, and 44% of values below quantification limits for total, pesticides, and PPCPs and additives sums, respectively, this method gave significant results considering the whole dataset. Even though the negative binomial regression integrates these values in the analysis, a higher proportion of quantified data might provide more robust results because there would be more ranks in the data, as data below detection limits all have the same rank. However, this is partly due to the design of the data collection study encompassing lakes with little to no anthropogenic impact in their watersheds. Such ecosystems are not usually included in contamination studies, overlooking an important side of lakes' diversity.

3.7.3. Recommendations for Canadian lakes

Considering all the observations and obtained results, lakes with high proportions of anthropogenic land use in their watersheds, such as agriculture or urbanization, should be monitored for TrOCs. Indeed, more intense contamination has been found in highly anthropogenic areas, with concentrations that could pose a high risk to aquatic organisms in those lakes (Lahens et al., 2024). It is thus important to reduce anthropogenic impacts by prioritizing our efforts to limit TrOCs discharge and entry into aquatic environments.

Among lakes sampled in the Lake Pulse survey, only 290 were analyzed for TrOCs. Within the other 369 sampled lakes, some sustain high human pressures and could be at risk of exhibiting significant trace organic contamination (Annexe 2 **Tableau 22**). Of these lakes, the 10 lakes with the most anthropogenic watersheds are listed in **Tableau 5**. An unnamed lake in Ontario, Barnaby Lake, BC, and Lakes Bissett and Banook, NS, have highly urbanized watersheds. They could be expected to display high detections and concentrations of TrOCs, and more specifically PPCPs and additives. Wilson and Jumping Lakes, SK, as well as unnamed lakes in Saskatchewan, and Alberta have high fractions of their watersheds allocated to agriculture and could exhibit elevated pesticide concentrations. Two other lakes, Laxton Lakes, BC, and an unnamed lake in Ontario, could contain a mixture of TrOCs as their watersheds are used for both agriculture and urbanization. The Ontarian lake also has a WWTP on its watershed and has high odds of having great concentrations of PPCPs and additives.

Tableau 5. Lakes with the highest anthropogenic land use fractions within the Lake Pulse survey but not sampled for TrOCs.

Lake name	Latitude	Longitude	Agricultural fraction	Urban fraction	Presence of WWTPs
NA, ON	43.747219	-79.735133	0%	96%	No
Burnaby Lake, BC	49.243327	-122.946538	0%	92%	No
Bissett Lake, NS	44.655567	-63.469954	0%	89%	No
Lake Banook, NS	44.692693	-63.553527	0%	87%	No
Wilson Lake, SK	51.31598	-102.877558	84%	1%	No
NA, ON	42.874465	-82.183136	73%	11%	Yes
Jumping Lake, SK	52.855734	-105.447346	80%	3%	No
NA, SK	50.392956	-109.488101	79%	2%	No
NA, AB	53.547125	-111.801211	76%	4%	No
Laxton Lake, BC	49.008293	-122.35186	50%	30%	No

3.8. Conclusions

In the present study, disparity in the distribution of TrOCs in Canadian lakes was highlighted and the drivers of these variations were verified in a large-scale setting. Urban and agriculture land use fractions and the presence of WWTPs in lakes' watersheds were confirmed as significant factors and showed variable effects depending on the contaminant classes. Moreover, some lakes with high risk of contamination have been highlighted and could be considered for future monitoring.

Future studies could include additional anthropogenic activities such as forestry, mining or oil drilling, more detailed land use categories, or other types of contamination sources such as atmospheric depositions. Nonetheless, these results can help policy makers make informed decisions on watershed management. Overall, these results highlight the importance of a few factors in these highly complex ecosystems, thus helping to focus on a limited number of useful variables for identifying potential contamination risks. Research efforts could focus on improving wastewater treatment and hospital effluents pre-treatment, while better management of pesticide use and mitigation efforts on leaching at the watershed scale could also improve lake water quality in agricultural as well as urbanized areas.

3.9. CRediT authorship contribution statement

Lisa Lahens: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing - Original Draft, Writing - Review & Editing, Visualization. **José A. Correa**: Methodology, Validation, Writing - Review & Editing. **Hubert Cabana**: Writing - Review & Editing. **Yannick Huot**: Resources, Writing - Review & Editing, Funding acquisition. **Pedro A. Segura**: Conceptualization, Resources, Writing - Review & Editing, Supervision.

3.10. Declaration of competing interest

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

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3.12. Appendix A. Supplementary data

Supporting information relating to this article are available online and refer to detailed data, model scripts and validation results (Annexe 2).

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CONCLUSION GÉNÉRALE

Les lacs sont des écosystèmes ayant une grande importance écologique, aussi bien du point de vue de l'environnement que de celui des services rendus à l'humain. En effet, c'est une source d'eau potable, d'énergie et de biodiversité, mais aussi de bien-être psychologique et spirituel, qui a une importance particulière au Canada qui est le pays ayant le plus de lacs sur son territoire. Or ces écosystèmes sont peu étudiés en comparaison des rivières et autres cours d'eau (Meyer et al., 2019).

Cette recherche est la première à avoir apporté des informations sur la distribution d'une cinquantaine de contaminants organiques de classes et de sources différentes dans les lacs à l'échelle d'un pays, d'autant plus dans un territoire aussi étendu. Un grand nombre de ces lacs présente une contamination suffisamment élevée pour faire courir un risque potentiel aux organismes aquatiques vivant dans leurs eaux. Cette étude a été une opportunité unique d'étudier les facteurs de contamination avec des données provenant de bassins versants hétérogènes et représentatifs d'une grande variété de lacs, mettant en avant l'importance de l'agriculture, de l'urbanisation et de la présence de STEP sur la détection et la quantification de TrOCs. La collaboration avec un statisticien a permis de mettre en œuvre un modèle novateur dans le domaine. En effet, l'application d'un modèle de régression logistique ordinale a permis de prendre en compte les résultats sous les limites de détection, qui sont très fréquents dans l'analyse de la contamination environnementale. Le script permettant l'application de ce modèle a été partagé afin de permettre à d'autres chercheurs d'utiliser cette méthodologie. La mise en évidence de quelques variables de grande importance dans ces écosystèmes complexes pourrait permettre de prédire le risque de contamination des lacs pour lesquels les données sur ces variables sont disponibles, ce qui pourrait aider à mettre en lumière des régions à risque de contamination, notamment en cas de changements dans l'utilisation du sol des bassins versants. Les résultats de cette thèse pourront donc servir dans la poursuite de l'étude de la contamination des lacs canadiens, mais également à une échelle plus globale dans la gestion des bassins versants lacustres et des sources de contamination. En outre, ces résultats soulignent l'importance d'une meilleure gestion des pratiques d'utilisation des sols et peuvent être utilisés par les autorités locales pour prendre des décisions éclairées sur la priorisation des lacs pour une recherche plus poussée des sources de contamination et des efforts de conservation. Afin d'aller plus loin dans la compréhension des impacts humains sur les lacs, une étude approfondie de certains lacs à risque pourrait être menée à l'échelle de leurs bassins versants en incluant l'impact d'autres activités anthropiques telles que la sylviculture, l'exploitation minière ou le forage pétrolier, des catégories d'utilisation des sols plus détaillées ou d'autres types de sources de contamination telles que les dépôts atmosphériques.

Cependant, cette étude comporte certaines limitations. En effet, des échantillons uniques pour chaque lac ont été prélevés en été, donnant lieu à une étude ponctuelle de la contamination. Dans le cadre d'une étude à si grande échelle, il n'était pas possible d'effectuer un suivi saisonnier sur plusieurs centaines de lacs à travers le pays. Néanmoins, les lacs subissent de fortes variations saisonnières qui pourraient impacter les concentrations de TrOCs ainsi que leur distribution dans la colonne d'eau des lacs (Baldwin et al., 2016; Helm et al., 2012). Pour combler cette limite, un suivi temporel de quelques lacs ayant des caractéristiques de bassins versants différentes permettrait de mettre en perspectives les résultats obtenus. Une autre limitation est liée à la méthode d'analyse. En effet, afin de pouvoir analyser des composés de différentes classes, une méthode d'analyse multi-résidus a été développée. Cependant, l'analyse simultanée de composés très variables nécessite de faire des compromis sur les limites de détection et de quantification pour chaque composé. De plus, certains composés nécessitant l'utilisation d'appareils analytiques différents, tels que les PFAS ou les PAH, n'ont pas pu être inclus dans cette méthode. Leur analyse aurait pu mettre en évidence d'autres lacs à risque. Une solution permettant d'avoir de meilleures limites en incluant une plus grande variété de contaminants aurait été de développer plusieurs méthodes d'analyse en fonction des familles de composés. Ce procédé aurait toutefois nécessité plus de moyens et plus de temps, notamment avec un si grand nombre d'échantillons à analyser. Il était donc nécessaire de mettre en place une méthode unique pour couvrir un large spectre de sources potentielles tout en limitant le temps d'analyse afin d'avoir une meilleure idée de la contamination globale de ces nombreux lacs.
De plus, une importante limitation est l'utilisation d'une méthode d'analyse ciblée, qui ne permet donc de trouver que les composés que l'on cherche. Bien que la sélection des composés ait été faite en prenant compte de plusieurs paramètres tels que leur omniprésence dans l'environnement ou leur potentielle toxicité, ce type d'analyse peut manquer de mettre en évidence des contaminants importants n'ayant pas été inclus dans la liste des cibles. Néanmoins, les analyses non-ciblées ont un coût plus élevé et un temps d'analyse plus long. Il est donc difficile de mettre en place de telles méthodes pour un grand nombre d'échantillons. Il serait cependant intéressant d'effectuer des analyses non-ciblées sur une sélection de lacs plus à risque de contamination.
Enfin, l'évaluation du risque environnemental, bien que suivant les directives de la Commission Européenne, comporte des limitations puisqu'elle ne prend en compte que l'exposition des organismes aquatiques aux différents TrOCs et le danger posé par ces contaminants pour des espèces modèles. Une

évaluation plus complète pourrait être effectuée en étudiant de façon plus approfondie la biodiversité des lacs échantillonnés ainsi que la structure des communautés. Ces données pourraient être mises en relation avec les résultats de contamination organique afin d'évaluer l'impact réel de ces TrOCs sur le milieu.

Les travaux effectués dans cette thèse permettent donc de combler le manque d'information sur la contamination des lacs en apportant des données sur la présence et les niveaux de concentration de TrOCs dans les eaux de près de 300 lacs à travers le Canada, mettant en évidence l'omniprésence de la contamination anthropique dans ces lacs. En plus de fournir une base pour un potentiel suivi de la qualité des lacs canadiens, ces données de concentrations ont permis d'estimer le risque encouru par les organismes lacustres ainsi que de mettre en évidence les principaux facteurs menants à cette contamination. De plus, des lacs canadiens non échantillonnés ayant des bassins versants fortement anthropisés ont été recommandés pour un potentiel suivi. Les résultats seront aussi rendus publiques afin d'informer les citoyens de l'état de santé des lacs canadiens et permettre une conscientisation de l'impact humain sur l'environnement. Les données obtenues peuvent également contribuer à améliorer les politiques gouvernementales de protection des écosystèmes aquatiques. En effet, ce projet entre dans les ambitions de la stratégie québécoise de l'eau 2018-2030 dont une des orientation est d'« acquérir et partager les meilleures connaissances sur l'eau ». L'obtention de ces données sur la contamination des lacs, dont 51 se trouvent au Québec, ainsi que leur publication et leur future mise à disposition au grand public participent à ces objectifs (MDDELCC, 2018). Certains résultats ont déjà été communiqués à Agriculture Canada lors d'une réunion de discussion sur l'impact de l'agriculture sur les lacs, et un mémoire sur l'impact potentiel des pesticides dans les milieux aquatiques du Québec a été soumis dans le cadre de la consultation générale sur les impacts des pesticides sur la santé publique et l'environnement de la Commission de l'agriculture, des pêcheries, de l'énergie et des ressources naturelles du Québec.

Enfin, cette recherche s'inscrit dans un projet multidisciplinaire, Lake Pulse, pour lequel un grand nombre de variables a été étudié. Les résultats obtenus au cours de ce doctorat pourront donc être mis en relation avec les données acquises par d'autres groupes de recherche. L'impact des antibiotiques dans l'eau des lacs canadiens sur la présence de gènes de résistance aux antibiotiques pourrait par exemple être évalué. Plus largement, les données de présence et de concentrations des TrOCs pourraient être mise en lien avec les structures de communautés de phytoplancton et de zooplancton dans les lacs afin de mettre en évidence de potentiels effets des mélanges de contaminants sur ces organismes.

4.1. Références

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ANNEXE 1. INFORMATIONS SUPPLÉMENTAIRES DU CHAPITRE 2

A1.1. Detailed Sample Preparation

After thawing, samples were filtered on 1.2-µm GFF, followed by 0.45-µm membrane filters. The pH of the filtered samples was adjusted to 6.5 with HCl or NaOH as the lakes of interest had highly variable pH, ranging from 4 to 10, and 200 mg/L of EDTA was added to prevent contaminants from complexing metal ions. For the 2017 campaign, 200 mL sub-samples were concentrated by a factor of 500 by solid phase extraction (SPE) using Strata-X cartridges (Phenomenex, 200 mg, 6 mL). These cartridges contain a surface-modified co-polymer of styrene-divinylbenzene as solid phase and were used for extraction of samples. Cartridges were first conditioned with 5 mL of ACN-MeOH (1:1, *v/v*) and 5 mL of Milli-Q water (pH adjusted to 6.5). The samples were then loaded at a flow rate of 3 mL/min. Cartridges were washed with 2 × 5 mL of pH 6.5 Milli-Q water and vacuum dried for 10 min before the analytes were eluted with 2 × 2.5 mL of ACN-MeOH (1:1, *v/v*) in glass tubes. Deuterated internal standards were added, and samples were evaporated to dryness under a gentle nitrogen stream in a sand bath at 35°C before reconstitution in H_2O-MeOH (80:20, *v/v*). The 2018 and 2019 samples were extracted with a Gilson GX-274 ASPEC automated SPE system (Gilson Inc., Middleton, WI, USA) using the same extraction parameters than the 2017 samples except the cartridge drying step, which was done under a nitrogen flow at 25 psi for 5 min and the drying of the samples after addition of the deuterated compounds in an evaporation module set to 35°C.

A1.2. Analytical Apparatus

Quantification of contaminants was done using a liquid chromatography-triple quadrupole mass spectrometry (LC-QqQMS) system from Waters (Milford, MA, USA). The LC-QqQMS was composed of an Acquity Binary Solvent Manager system coupled to a triple quadrupole mass spectrometer: a Xevo TQ for the 2017 samples and a Xevo TQ-S micro for the 2018 and 2019 samples. Chromatographic separation was achieved using an Acquity UPLC HSS T3 column (50 mm × 2.1 mm i.d., 1.8 µm particle size) made by Waters. Mobile phase flow rate was 0.5 mL/min, and the sample injection volume was set to 5 µL.

A1.3. Method Validation

Extraction recoveries (ER) were assessed by spiking four replicates before and four replicates after extraction using the following equation:

$$ER(\%) = \frac{Mean\ response_{Before}}{Mean\ response_{After}} \times 100 \qquad [A1.1]$$

The development and validation of the analytical methods were performed on pristine lake samples collected in a nearby lake (Lake Gale, QC) using two sets of seven replicates spiked at 20 and 200 ng/L. Matrix blank subtraction was done on controls and calibration points to account for potential contamination. The range of validation results for all methods are presented in Annexe 1 **Tableau 10** and details are given in Annexe 1 **Tableau 11**. Seven-point calibration curves were produced using ordinary linear least squares regression analysis. The calibrations usually ranged from 0 to 500 ng/L but, for some compounds, curves went up to 7 µg/L occasionally. LOQ was defined as the smallest calibration point with a deviation < 20% and LOD was calculated as follows:

$$LOD = \frac{LOQ}{3.3} \qquad [A1.2]$$

These values were calculated for each calibration curve. The ranges of LOD and LOQ are shown in Annexe 1 **Tableau 10**.

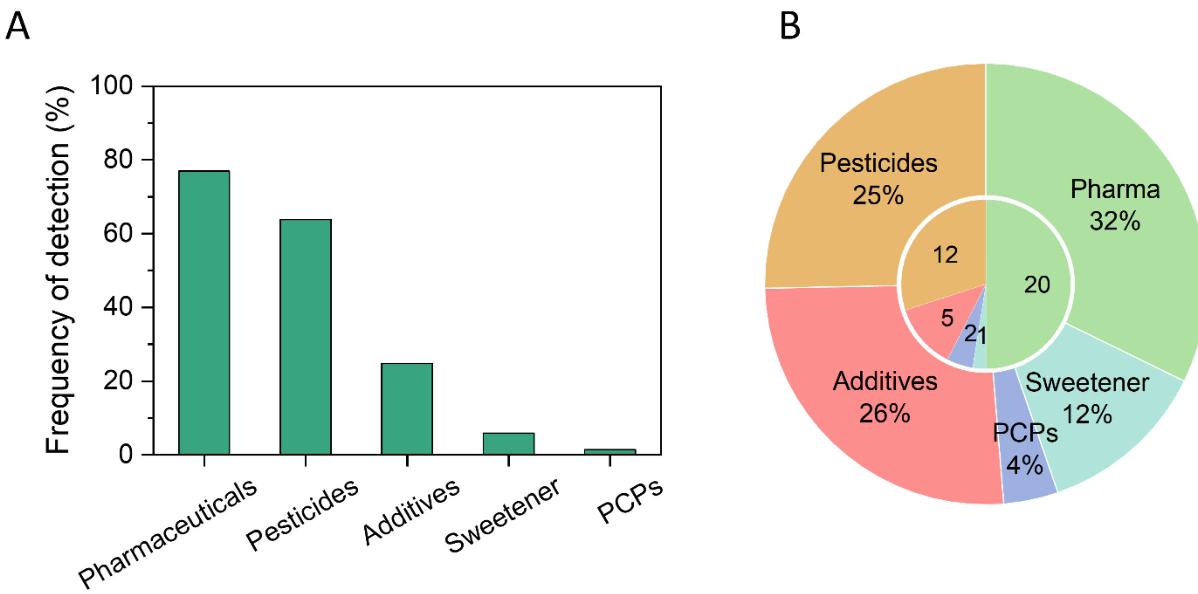

Figure 11. Statistics of contaminants presence by usage class: A) frequency of detection of different classes of contaminants, and B) percentage of the total summed concentration (outer circle) and number of quantified compounds (inner circle) of target TrOCs by usage category.

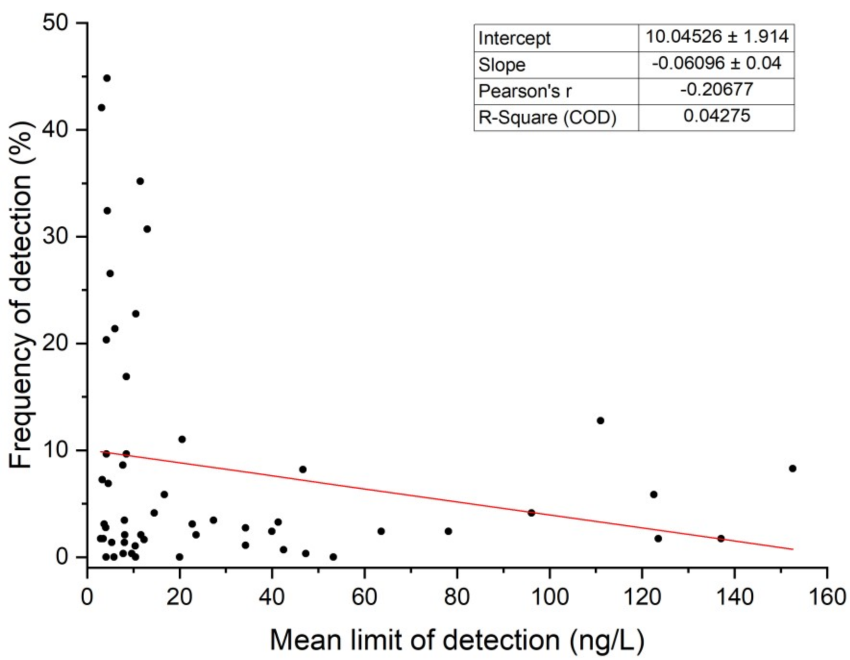

Figure 12. Frequencies of detection of the targeted contaminants as a function of the mean of all method detection limits for that compound. The linear model is not significant with $p = 0.1336$.

Tableau 6. Information on the sampled lakes.

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
04-572	Nataiinlaii Gwint'ii Van, NT	67.332779	-134.844522	2019-07-31	0.5	moderate	3	4
04-577	Unnamed lake, NT	68.344096	-133.705674	2019-08-05	0.1	high	3	4
04-579	Campbell Lake, NT	68.232838	-133.44037	2019-08-07	35.7	low	3	4
04-580	Unnamed lake, NT	67.46993	-134.72108	2019-07-30	0.7	moderate	3	4
04-583	Unnamed lake, NT	67.442962	-134.511877	2019-08-02	0.3	low	3	4
04-609	Heart Lake, NT	60.836026	-116.656975	2019-07-30	2.5	low	3	4
04-612	Unnamed lake, NT	60.721344	-114.996196	2019-07-27	0.7	moderate	3	4
04-613	Unnamed lake, NT	60.813148	-114.589631	2019-07-28	0.1	low	3	4
04-614	Unnamed lake, NT	60.53671	-114.568217	2019-07-29	0.2	low	3	4
04-615	Pine Lake, AB	59.534902	-112.216745	2019-08-01	3.0	low	3	4
04-616	Unnamed lake, AB	59.944209	-111.853586	2019-08-02	0.4	low	3	4
04-617	Unnamed lake, NT	60.596644	-116.262865	2019-08-03	0.3	low	3	4
04-618	Chan Lake, NT	61.890891	-116.542274	2019-08-07	0.8	low	3	4
04-619	Unnamed lake, NT	62.556145	-116.433386	2019-08-05	0.1	low	3	4
04-623	Parker Lake, BC	58.822968	-122.901342	2019-08-12	0.6	low	3	4
04-624	Unnamed lake, BC	58.623863	-122.693773	2019-08-13	0.2	low	3	4
06-083	Lac Saint-Antoine, QC	47.534528	-70.232947	2017-08-19	0.3	moderate	3	1
06-085	Lac Nairne, QC	47.685661	-70.349677	2017-08-18	2.5	moderate	3	2
06-102	Lac Froget, QC	50.732795	-71.771475	2017-08-17	3.1	high	3	1
06-103	Lac à la Croix, QC	48.395105	-71.773708	2017-08-22	0.7	high	3	1
06-104	Lac Vouzier, QC	48.407729	-71.78678	2017-08-20	0.1	high	3	2

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
06-126	Lac Paula, QC	48.993311	-74.029437	2017-08-25	0.6	high	3	1
06-127	Lac Duminy, QC	48.987717	-74.06735	2017-08-26	0.2	moderate	3	1
06-128	Lac des Seigneurs, QC	45.862265	-74.115675	2017-07-14	0.2	high	3	3
06-129	Lac Winsch, QC	50.032073	-74.221815	2017-08-24	1.3	low	3	1
06-130	Lac Marois, QC	45.850978	-74.131647	2017-07-13	1.0	high	3	2
06-132	Lac Sainte-Marie, QC	45.958676	-74.266369	2017-07-16	2.1	low	3	3
06-133	Lac Duhamel, QC	46.142222	-74.638943	2017-07-17	0.5	low	3	3
06-136	Lac Ouellette, QC	46.719648	-75.437263	2017-08-28	0.6	high	3	2
06-137	Lac des Îles, QC	46.450343	-75.551099	2017-08-29	16.7	low	3	1
06-139	Lac Vert, QC	45.902448	-75.605782	2017-08-30	0.3	high	3	1
06-158	Lac Merlin, QC	48.893947	-76.881395	2017-08-21	0.2	moderate	3	3
06-174	Lac Wasi, ON	46.138734	-79.225947	2017-09-02	6.9	moderate	3	1
06-200	Unnamed lake, ON	48.238266	-81.040785	2017-08-13	0.1	high	3	3
06-217	Borden Lake, ON	47.836499	-83.286417	2017-08-12	16.6	low	3	2
06-220	Bright Lake, ON	46.272242	-83.30755	2017-08-10	12.5	moderate	3	1
06-221	Sandbar Lake, ON	47.835215	-83.348195	2017-08-11	0.3	moderate	3	2
06-254	Little Rushy Pond, NL	48.949782	-55.693761	2018-08-08	0.4	high	3	4
06-258	Hogans Pond, NL	47.583623	-52.852923	2018-08-12	0.6	high	3	4
06-261	Big Otter Pond, NL	47.399963	-53.04748	2018-08-13	0.7	moderate	3	4
06-274	Rabbit Lake, ON	49.787686	-94.463764	2018-07-05	0.9	high	3	4
06-291	Camp Lake, MB	55.128342	-101.103371	2018-07-26	0.5	high	3	4
06-293	Liz Lake, MB	55.487013	-98.050248	2018-07-28	2.0	moderate	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
06-304	L226, ON	49.688665	-93.747517	2018-08-14	0.2	low	3	4
06-315	Johnson's Lake, ON	49.711052	-83.684753	2018-08-28	0.4	high	3	4
07-002	Larkins Pond, PE	46.421872	-62.428828	2017-07-21	0.3	low	3	3
07-003	Lake Verde, PE	46.23885	-62.885457	2017-07-20	0.2	high	3	3
07-005	Long Lake, NS	45.902727	-64.155071	2017-07-23	1.0	low	3	2
07-006	Collins Lake, NB	46.110489	-64.150702	2017-07-24	0.7	low	3	1
07-007	Blair Lake, NS	45.799248	-64.210287	2017-07-29	0.5	high	3	1
07-008	Morice Pond, NB	45.931138	-64.368213	2017-07-28	1.5	moderate	3	2
07-009	Black River Lake, NS	44.930654	-64.418764	2017-08-13	7.5	low	3	2
07-010	Gaspereau Lake, NS	44.953072	-64.563372	2017-08-11	22.0	low	3	3
07-013	Colwell Round Lake, NS	44.845846	-64.598487	2017-08-14	0.1	low	3	2
07-015	Spectacle Lake, NS	44.259495	-64.610661	2017-08-18	1.0	moderate	3	2
07-020	Unnamed lake, NB	47.58432	-64.955436	2017-07-17	0.2	low	3	3
07-022	Lake Mulgrave, NS	44.496459	-65.471246	2017-08-09	14.5	low	3	2
07-023	Goose Lake, NS	43.603816	-65.527065	2017-08-19	0.6	low	2	1
07-024	Barrington Lake, NS	43.612856	-65.573869	2017-08-20	1.6	low	3	2
07-026	Crocker Lake, NB	46.89332	-65.727509	2017-07-15	0.8	low	3	2
07-029	Ritchie Lake, NB	45.415706	-65.96754	2017-08-05	0.2	high	3	3
07-030	Lily Lake, NB	45.290432	-66.057534	2017-08-04	0.1	high	3	3
07-031	Lac Innocent, NS	44.273871	-66.075345	2017-08-25	0.1	high	3	1
07-033	Napadogan Lake, NB	46.416483	-66.943837	2017-08-02	0.3	moderate	3	2
07-035	McKendrick Lake, NB	46.852777	-66.361613	2017-07-23	0.8	low	3	1

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
07-037	Moose Lake, NB	45.448897	-66.47072	2017-08-06	0.7	low	3	1
07-040	McDougall Lake, NB	45.324348	-66.774486	2017-08-27	3.1	low	3	1
07-052	Davidson Lake, NB	45.93731	-67.158731	2017-08-30	2.1	moderate	3	1
07-228	Fletchers Lake, NS	44.842279	-63.611864	2018-07-20	1.0	high	3	4
07-229	First Lake, NS	44.770926	-63.661881	2018-07-23	0.9	high	3	4
07-230	Fenerty Lake, NS	44.831734	-63.719825	2018-07-15	0.7	moderate	3	4
07-231	Moore Lake, NS	44.961142	-63.757733	2018-07-08	0.1	moderate	3	4
07-236	Governor Lake, NS	44.642918	-63.701964	2018-07-16	0.4	high	3	4
07-237	Five Island Lake, NS	44.664418	-63.806279	2018-07-22	1.4	moderate	3	4
07-243	Lake Charles, NS	44.722618	-63.551142	2018-07-14	1.4	high	3	4
07-247	MacPherson Lake, NS	45.433201	-61.420529	2018-07-30	0.9	moderate	3	4
07-248	Shepherd Lake, NS	45.522507	-61.560681	2018-07-29	0.3	low	3	4
07-250	Lake Ainslie, NS	46.132703	-61.185324	2018-08-02	58.8	low	3	4
07-251	Unnamed lake, NS	46.170166	-60.014699	2018-08-03	0.2	moderate	3	4
08-097	Lac Saint-Augustin, QC	46.749625	-71.392292	2017-08-26	0.7	high	3	1
08-118	Lac Saint-Paul, QC	46.304437	-72.476705	2017-08-20	2.9	high	3	1
08-134	Loch Garry, ON	45.254158	-74.709611	2017-09-11	3.9	low	3	1
08-135	Lac Georges, ON	45.605917	-74.97345	2017-09-12	0.6	moderate	3	1
08-145	Cedar Lake, ON	44.418957	-76.399919	2017-07-21	0.2	moderate	2	3
08-147	Mississippi Lake, ON	45.035782	-76.184422	2017-09-09	25.2	low	3	2
08-151	Devil Lake, ON	44.577578	-76.440764	2017-07-16	11.0	low	3	2
08-154	Lacs Healy, QC	45.844902	-76.653917	2017-09-03	0.1	low	3	1

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
08-155	Varty Lake, ON	44.391856	-76.813362	2017-07-22	6.3	moderate	3	2
08-160	Stoco Lake, ON	44.473281	-77.28962	2017-07-25	5.8	low	3	2
08-164	Stevenson Lake, ON	44.274574	-77.910623	2017-07-29	0.2	moderate	3	3
08-166	Rice Lake, ON	44.1779	-78.179116	2017-07-28	99.7	low	3	2
08-168	Sturgeon Lake, ON	44.474286	-78.687323	2017-09-06	46.4	moderate	3	1
08-175	Sunova Lake, ON	43.194682	-81.02096	2017-08-18	0.3	high	3	3
08-177	Lake St. John, ON	44.687068	-79.325113	2017-09-04	6.8	moderate	3	1
08-179	Wilcox Lake, ON	43.949049	-79.436034	2017-08-01	0.6	high	3	3
08-180	Eversley Lake, ON	43.957772	-79.500823	2017-08-03	0.1	high	3	2
08-182	Little Lake, ON	44.426424	-79.671388	2017-08-24	2.4	high	3	1
08-184	Heart Lake, ON	43.740539	-79.795426	2017-08-05	0.2	moderate	3	1
08-186	Lake Niapenco, ON	43.105298	-79.850926	2017-08-20	1.9	high	3	1
08-187	Farden Lake, ON	44.276532	-80.690839	2017-07-24	0.1	moderate	3	2
08-190	Pinehurst Lake, ON	43.269235	-80.390052	2017-08-21	0.1	moderate	3	1
08-192	Green Lake, ON	43.839801	-80.008756	2017-08-08	0.1	high	3	2
08-193	Dankert Lake, ON	44.205289	-81.052711	2017-08-12	0.2	high	3	2
08-202	Boat Lake, ON	44.725929	-81.227067	2017-07-23	5.6	moderate	3	3
08-205	Gillies Lake, ON	45.204954	-81.326861	2017-07-28	2.2	high	3	4
08-206	Cyprus lake, ON	45.231341	-81.531897	2017-07-31	0.8	low	3	2
08-208	Clam Lake, ON	44.072133	-81.413948	2017-08-13	0.4	high	3	1
08-210	Unnamed lake, ON	43.575686	-81.661215	2017-08-14	0.2	high	3	1
08-211	Sucker Lake, ON	45.723555	-81.878484	2017-08-08	2.3	low	3	2

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
08-212	Pike Lake, ON	45.87402	-81.985603	2017-08-07	2.3	low	3	3
08-214	Unnamed lake, ON	44.007233	-78.029579	2017-09-07	0.1	moderate	3	1
08-218	Falls Lake, ON	45.898543	-83.106275	2017-08-04	1.1	low	3	1
08-219	Lac Mccord, QC	45.693047	-76.46411	2017-09-04	0.1	low	3	2
09-286	Nut Lake, SK	52.343345	-103.706775	2018-07-20	18.0	low	3	4
09-292	Egg Lake, MB	54.37048	-101.457133	2018-07-27	30.6	low	3	4
09-297	Devils Lake, MB	52.398538	-98.912922	2018-08-02	0.5	low	3	4
09-369	Unnamed lake, AB	53.542469	-117.070949	2018-07-10	0.1	moderate	3	4
09-370	Fickle Lake, AB	53.449373	-116.779894	2018-07-12	3.6	low	3	4
09-375	Lac la Nonne, AB	53.939715	-114.321366	2018-07-19	12.5	moderate	3	4
09-385	Jones Lake, AB	55.391549	-119.003383	2018-07-30	1.3	moderate	3	4
09-392	Unnamed lake, AB	54.894823	-112.316588	2018-08-09	0.2	moderate	3	4
09-393	Claude Lake, AB	54.793696	-111.909232	2018-08-10	0.7	moderate	3	4
09-397	Upper Thérien Lake, AB	53.965983	-111.293303	2018-08-14	8.1	high	3	4
09-409	Falcon Lake, MB	50.50351	-99.953734	2018-07-03	0.3	high	3	4
09-411	Little Jackfish Lake, MB	50.47746	-100.074738	2018-07-02	1.6	high	3	4
09-427	Poplar Ridge Lake, SK	53.92408	-107.702852	2018-07-26	0.1	low	3	4
09-428	Green Lake, SK	54.09185	-107.6804	2018-07-27	27.8	low	3	4
09-430	Unnamed lake, SK	54.993693	-108.344502	2018-07-28	0.4	low	3	4
09-449	Unnamed lake, MB	51.439365	-101.42674	2018-08-24	0.1	high	3	4
09-450	Meeting Lake, SK	53.188577	-107.657995	2018-08-19	10.8	moderate	3	4
09-596	Pigeon Lake, AB	53.024412	-114.059639	2019-07-07	94.8	low	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
09-602	Willow Lake, AB	56.462556	-111.162013	2019-07-16	25.8	low	3	4
09-603	Kearl Lake, AB	57.292089	-111.238794	2019-07-15	5.1	high	3	4
09-604	Saskatoon Lake, AB	55.217558	-119.091379	2019-07-20	8.0	high	3	4
09-607	Kakut Lake, AB	55.628822	-118.528498	2019-07-21	3.6	moderate	3	4
10-283	Bennet Lake, SK	49.771608	-102.458801	2018-07-15	0.3	low	3	4
10-284	Unnamed lake, SK	49.939593	-102.451854	2018-07-16	0.9	moderate	3	4
10-301	North Shoal Lake, MB	50.470346	-97.650318	2018-08-08	31.9	low	3	4
10-346	Beaverdam Lake, AB	49.084553	-113.606695	2018-08-11	0.7	low	3	4
10-349	Unnamed lake, AB	51.591071	-113.753331	2018-08-16	0.3	high	3	4
10-352	Namaka Lake, AB	50.939508	-113.232916	2018-08-19	6.4	moderate	3	4
10-355	Lac Pelletier, SK	49.986537	-107.934986	2018-08-23	3.0	high	3	4
10-356	Fife Lake, SK	49.217733	-105.851938	2018-08-24	29.4	moderate	3	4
10-357	Unnamed lake, SK	50.21254	-103.584538	2018-08-25	0.5	high	3	4
10-358	Little Kenosee Lake, SK	49.83338	-102.332239	2018-08-28	1.5	low	3	4
10-359	Alkali Lake, SK	49.813839	-102.397321	2018-08-29	0.1	low	3	4
10-387	Driedmeat Lake, AB	52.85999	-112.74476	2018-08-24	12.1	high	3	4
10-388	Boag Lake, AB	53.518523	-113.219575	2018-08-04	0.9	moderate	3	4
10-389	Skaro Lake, AB	53.924854	-112.714336	2018-08-05	0.4	high	3	4
10-398	Birch Lake, AB	53.316011	-111.589784	2018-08-17	8.2	moderate	3	4
10-399	Thomas Lake, AB	53.110315	-111.699187	2018-08-18	5.1	high	3	4
10-400	Macklin Lake, SK	52.320211	-109.95027	2018-08-26	0.8	moderate	3	4
10-402	Miquelon Lakes, AB	53.256011	-112.912167	2018-08-21	7.7	low	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
10-406	Red Deer Lake, AB	52.721431	-113.060046	2018-08-25	21.1	moderate	3	4
10-412	Shoal Lake, MB	50.388326	-100.626474	2018-07-06	4.8	high	3	4
10-416	Unnamed lake, SK	52.239307	-103.947363	2018-07-10	0.6	moderate	3	4
10-433	Unnamed lake, SK	52.635863	-108.567479	2018-08-02	0.2	high	3	4
10-435	Manitou Lake, SK	52.738966	-109.652625	2018-08-05	81.2	moderate	3	4
10-438	Crookshanks Lake, SK	51.860856	-109.311232	2018-08-04	0.3	high	3	4
10-444	Murray Lake, SK	53.037189	-108.278678	2018-08-16	11.6	moderate	3	4
10-445	Schmidt Lake, SK	52.670482	-109.211384	2018-08-14	0.5	moderate	3	4
10-452	Lytwyns Lake, MB	51.597074	-99.849842	2018-08-26	0.3	low	3	4
10-453	Unnamed lake, SK	52.671989	-108.310133	2018-08-13	0.5	low	3	4
10-597	Muir Lake, AB	53.623604	-113.95501	2019-07-08	0.3	moderate	3	4
10-598	Astotin Lake, AB	53.678768	-112.853935	2019-07-09	5.5	low	3	4
11-342	Whiteswan Lake, BC	50.144066	-115.474903	2018-08-06	4.1	low	3	4
11-470	Dunalter Lake, BC	54.470753	-126.755752	2019-07-14	0.2	moderate	3	4
11-478	Ferguson Lake, BC	54.037199	-122.84621	2019-07-24	0.2	low	3	4
11-480	Hobson Lake, BC	53.580617	-124.73149	2019-07-27	0.6	low	3	4
11-481	Fish Lake, BC	53.598731	-124.889241	2019-07-28	0.4	low	3	4
11-482	Unnamed lake, BC	53.875399	-124.755299	2019-07-29	0.1	low	3	4
11-483	Co-op Lake, BC	54.186152	-125.428674	2019-07-30	0.3	low	3	4
11-484	Unnamed lake, BC	53.977642	-125.898396	2019-08-01	0.3	high	3	4
11-486	Unnamed lake, BC	53.9938	-125.981445	2019-08-03	0.2	moderate	3	4
11-487	Totem Pole Lake, BC	53.966067	-125.944652	2019-08-04	0.3	moderate	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
11-488	Tatalaska Lake, BC	53.938458	-125.907128	2019-08-05	1.6	high	3	4
11-489	Mollice Lake, BC	53.957671	-125.714231	2019-08-06	1.9	moderate	3	4
11-491	Chief Lake, BC	54.120806	-123.004988	2019-08-10	7.1	moderate	3	4
11-493	Unnamed lake, BC	54.008382	-123.135854	2019-08-12	0.1	moderate	3	4
11-495	Williams Lake, BC	52.118825	-122.075003	2019-08-16	6.8	low	3	4
11-499	Konni Lake, BC	51.473574	-123.887461	2019-08-20	5.6	low	3	4
11-503	Clearwater Lake, BC	52.011725	-125.008155	2019-08-25	2.1	low	3	4
11-508	Italia Lake, BC	51.83278	-120.388941	2019-07-04	1.3	moderate	3	4
11-509	Latremouille Lake, BC	51.491893	-120.341789	2019-07-05	0.8	low	3	4
11-541	Takatoot Lake, BC	55.120772	-125.196624	2019-08-20	7.2	moderate	3	4
11-546	Unnamed lake, BC	54.168663	-124.208658	2019-08-24	0.1	moderate	3	4
11-547	Naltesby Lake, BC	53.607447	-123.488697	2019-08-26	8.2	low	3	4
11-556	Sepa Lake, BC	51.734097	-121.352237	2019-07-07	0.2	high	3	4
11-557	Greeny Lake, BC	51.850947	-121.33891	2019-07-08	0.8	low	3	4
11-589	Tudyah Lake, BC	55.084266	-123.038133	2019-08-23	6.2	low	3	4
11-627	Huble Lake, BC	54.269426	-122.616376	2019-08-18	0.2	low	3	4
11-628	Dominion Lake, BC	54.450825	-122.695605	2019-08-19	0.7	low	3	4
11-629	Summit Lake, BC	54.277084	-122.678241	2019-08-20	12.9	low	3	4
11-630	Rainbow Lake, BC	52.996317	-123.60365	2019-08-23	0.7	moderate	3	4
11-633	Eight Mile Lake, BC	53.149595	-121.534574	2019-08-26	0.1	moderate	3	4
12-460	Mill Lake, BC	49.044741	-122.310811	2019-06-29	0.2	high	3	4
12-462	Deer Lake, BC	49.236235	-122.97178	2019-06-30	0.3	high	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
12-466	Lillooet Lake, BC	50.215541	-122.490763	2019-07-06	31.9	low	3	4
12-473	Kitseguecla Lake, BC	54.934739	-127.55412	2019-07-13	0.7	low	3	4
12-504	Chehalis Lake, BC	49.443092	-122.020294	2019-08-26	6.3	low	3	4
12-528	Clements Lake, BC	56.051194	-129.902444	2019-08-01	0.2	low	3	4
12-529	Jigsaw Lake, BC	55.83382	-128.830233	2019-08-02	0.7	low	3	4
12-530	Derrick Lake, BC	55.649296	-128.644809	2019-08-03	0.5	low	3	4
12-531	Lava Lake, BC	55.047296	-128.993528	2019-08-04	5.2	low	3	4
12-533	Unnamed lake, BC	55.445634	-129.353516	2019-08-10	0.1	low	3	4
12-534	Unnamed lake, BC	55.446225	-129.334999	2019-08-09	0.2	low	3	4
12-535	Unnamed lake, BC	55.4549	-129.36962	2019-08-08	0.4	low	2	4
12-537	Taltzen Lake, BC	54.9301	-127.513355	2019-08-13	0.1	low	3	4
12-636	Thetis Lake, BC	48.467719	-123.4691	2019-07-02	0.2	moderate	3	4
12-637	Lake Weston, BC	48.784186	-123.425023	2019-07-04	0.2	moderate	3	4
12-640	Buck Lake, BC	48.772668	-123.300605	2019-07-07	0.1	high	3	4
12-641	Magic Lake, BC	48.763747	-123.288779	2019-07-08	0.1	high	3	4
12-642	Langford Lake, BC	48.448187	-123.529102	2019-07-11	0.6	high	3	4
12-644	Kemp Lake, BC	48.379522	-123.780515	2019-07-13	0.2	moderate	3	4
12-646	Dougan Lake, BC	48.714643	-123.613474	2019-07-15	0.1	high	3	4
12-648	Quamichan Lake, BC	48.799616	-123.662384	2019-07-18	2.9	high	3	4
12-649	Fuller Lake, BC	48.908326	-123.720615	2019-07-19	0.2	high	3	4
12-651	Holden Lake, BC	49.103249	-123.829263	2019-07-21	0.4	moderate	3	4
12-653	Boomerang Lake, BC	49.17875	-124.155461	2019-07-27	0.1	moderate	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
12-654	Kennedy Lake, BC	49.062864	-125.491128	2019-07-25	65.3	low	3	4
12-655	Lowry Lake, BC	49.390048	-125.135344	2019-07-26	0.4	low	3	4
12-656	Enos Lake, BC	49.28058	-124.156487	2019-07-30	0.2	moderate	3	4
12-657	Green Lake, BC	49.230739	-124.060791	2019-07-28	0.1	moderate	3	4
12-658	Mohun Lake, BC	50.11693	-125.499304	2019-08-05	6.2	low	3	4
12-659	McCreight Lake, BC	50.301082	-125.642773	2019-08-03	2.8	low	3	4
12-662	Victoria Lake, BC	50.371904	-127.387341	2019-08-09	15.7	low	3	4
12-665	Zeballos Lake, BC	50.075821	-126.754403	2019-08-12	2.0	low	3	4
12-670	Diver Lake, BC	49.203775	-124.014124	2019-08-15	0.1	high	3	4
12-671	Brannen Lake, BC	49.214698	-124.055142	2019-08-16	1.1	moderate	3	4
12-672	Long Lake, BC	49.210988	-124.017319	2019-08-17	0.4	high	3	4
12-675	Horne Lake, BC	49.333971	-124.675007	2019-08-20	8.1	low	3	4
12-676	Cranby Lake, BC	49.695238	-124.507497	2019-08-23	0.4	moderate	3	4
12-677	Haslam Lake, BC	49.936063	-124.420312	2019-08-25	11.6	low	3	4
13-514	Upper Gnat Lake, BC	58.21637	-129.837323	2019-07-12	0.4	moderate	3	4
13-516	Good Hope Lake, BC	59.300477	-129.276001	2019-07-14	1.7	low	3	4
13-517	Second Wye Lake, YT	60.062737	-128.680252	2019-07-16	0.3	moderate	3	4
13-519	Watson Lake, YT	60.107692	-128.80679	2019-07-15	13.4	low	3	4
13-521	Lower McDonald Lake, BC	59.703067	-133.60852	2019-07-20	0.5	low	3	4
13-523	Bennett Lake, YT	60.118864	-134.842427	2019-07-23	91.1	low	3	4
13-525	Dezadeash Lake, YT	60.479871	-137.010954	2019-07-26	74.2	low	3	4
13-526	Pine Lake, YT	60.816837	-137.448355	2019-07-25	5.8	low	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
13-560	Allan Lake, BC	58.430235	-130.001241	2019-07-13	0.4	moderate	3	4
13-562	Unnamed lake, YT	60.080432	-128.745812	2019-07-15	0.2	moderate	3	4
13-563	Unnamed lake, YT	61.455652	-129.745014	2019-07-16	0.1	moderate	3	4
13-565	Jackfish Lake, YT	61.934997	-132.521865	2019-07-18	1.1	low	3	4
13-566	Fish Eye Lake, YT	62.191856	-133.474391	2019-07-20	0.4	moderate	3	4
13-568	Ethel Lake, YT	63.36165	-136.062415	2019-07-23	43.3	low	3	4
13-569	Minto Lake, YT	63.685626	-136.161694	2019-07-24	4.0	low	3	4
13-587	Unnamed lake, YT	60.450307	-134.265322	2019-08-17	0.1	low	3	4
14-585	Two Moose Lake, YT	64.735027	-138.365273	2019-08-12	0.1	high	3	4
17-038	Middle Peaked Mountain Lake, NB	46.736363	-66.519057	2017-07-14	0.1	moderate	3	3
17-043	Trousers Lake, NB	47.010988	-66.970229	2017-07-20	11.1	moderate	3	1
17-050	Lac des Indiens, QC	50.202718	-66.174349	2017-08-11	0.1	high	3	3
17-054	Eightmile Lake, NB	47.695087	-67.644799	2017-07-31	0.2	high	3	2
17-056	Lac Désiré, QC	48.674467	-67.734944	2017-08-03	0.4	low	3	3
17-059	Lac de Saint-Damase, QC	48.653185	-67.809428	2017-08-07	0.7	high	3	2
17-069	Lac Jerry, QC	47.429633	-68.785243	2017-07-12	5.8	low	3	3
17-070	Lac Témiscouata, QC	47.686757	-68.848319	2017-07-29	67.7	low	3	2
17-072	Grand lac Malobès, QC	48.269232	-68.860309	2017-08-05	1.7	moderate	3	2
17-075	Lac du Marin-à-Gouin, QC	48.075148	-69.106521	2017-08-07	0.2	moderate	3	2
17-076	Lac Dole, QC	47.645885	-68.944567	2017-07-30	0.3	moderate	3	3
17-079	Lac Morin, QC	47.627411	-69.533598	2017-07-24	6.0	low	3	3

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
17-081	Lac de l'Est, QC	47.186479	-69.561309	2017-07-25	7.5	low	3	2
17-084	Lac du Portage, QC	45.935951	-70.270307	2017-07-21	4.3	low	3	2
17-087	Lac Mailloux, QC	46.706288	-70.490235	2017-07-11	0.2	moderate	3	3
17-088	Lac Etchemin, QC	46.389586	-70.493319	2017-07-22	2.5	moderate	3	2
17-090	Lac Volet, QC	46.137727	-70.809582	2017-08-22	0.3	low	3	2
17-092	Lac Mégantic, QC	45.537896	-70.889698	2017-07-18	28.3	moderate	3	3
17-093	Lac des Trois Milles, QC	45.687963	-70.921142	2017-07-19	1.0	moderate	3	3
17-094	Lac Jolicoeur, QC	46.061894	-71.10254	2017-08-26	0.2	high	3	2
17-098	Lac à la Truite, QC	46.084538	-71.500559	2017-08-24	1.2	moderate	3	1
17-105	Petit lac Saint-François, QC	45.537697	-72.037776	2017-08-27	0.8	high	3	2
17-107	Étang Burbank, QC	45.780093	-72.00684	2017-08-30	0.5	high	3	1
17-109	Lac Magog, QC	45.305175	-72.041315	2017-07-10	11.0	high	3	3
17-110	Lac Crystal, QC	45.030153	-72.076808	2017-07-13	0.2	high	3	3
17-111	Lac Montjoie, QC	45.408466	-72.098912	2017-07-10	3.4	low	3	2
17-114	Lac Brompton, QC	45.433218	-72.144938	2017-07-10	11.2	low	3	3
17-117	Lac Memphrémagog, QC	45.026848	-72.246826	2017-07-14	99.0	moderate	3	2
17-119	Lac Brome, QC	45.247947	-72.514469	2017-07-10	15.1	moderate	3	3
17-123	Lac Bromont, QC	45.265997	-72.671014	2017-07-11	0.5	moderate	3	2
18-324	Kalamalka Lake, BC	50.172799	-119.327369	2018-07-13	25.4	high	3	4
18-335	Nicola Lake, BC	50.166411	-120.528153	2018-07-23	24.9	low	3	4
18-336	Munro Lake, BC	49.715267	-119.923178	2018-07-28	0.1	low	3	4
18-337	Skaha Lake, BC	49.410038	-119.585408	2018-08-02	19.5	moderate	3	4

Lake ID	Lake name	Latitude	Longitude	Sampling date	Lake size (km^2)	Human impact class	Number of replicates	Analytical method*
18-338	Green Lake, BC	49.302628	-119.57101	2018-07-30	0.1	moderate	3	4
18-339	Vaseux Lake, BC	49.288771	-119.531568	2018-07-31	2.8	high	3	4
18-505	Chapperon Lake, BC	50.202061	-120.055602	2019-06-30	4.3	low	3	4
18-552	Stake Lake, BC	50.512281	-120.47799	2019-07-02	0.2	high	3	4
18-553	Strachan Lake, BC	50.903978	-120.610563	2019-07-04	0.1	high	3	4
18-593	Pennask Lake, BC	49.995289	-120.136936	2019-07-02	9.4	low	3	4

* (1) Manual SPE followed by analysis on the Xevo TQ mass spectrometer before July 2018
(2) Manual SPE followed by analysis on the Xevo TQ mass spectrometer from July 2018
(3) Automatic SPE with analysis on the Xevo TQ mass spectrometer
(4) Automatic SPE with analysis on the Xevo TQ-S micro mass spectrometer
See Annexe 1 **Tableau 11** for validation details

Tableau 7. Details on the classification of the selected contaminants.

Class	Sub-class	Use	Compound	Stock solution solvent (*v/v*)
Pharmaceutical products	Stimulant	Stimulant	Caffeine	H_2O/MeOH (2/3)
		Stimulant metabolite (caffeine)	1,7-Dimethylxanthine (Paraxanthine)	Basic H_2O/MeOH (1/4)
		Stimulant metabolite (nicotine)	Cotinine	MeOH
	Analgesic	Non-steroidal anti-inflammatory	Salicylic acid	MeOH
			Naproxen	MeOH
		Antipyretic	Acetaminophen	MeOH
	Antibiotic	Human antibiotic	Azithromycin	MeOH
			Ciprofloxacin	Acidic H_2O/MeOH (3/2)
			Sulfamethoxazole	MeOH
		Mixed antibiotic	Lincomycin	MeOH
			Oxytetracycline	MeOH
			Tetracycline	MeOH
			Trimethoprim	H_2O/MeOH (1/9)
		Mixed antibiotic metabolite (erythromycin)	Anhydroerythromycin A	MeOH
		Veterinary antibiotic	Ceftiofur	MeOH
			Chlortetracycline	MeOH
			Sulfamethazine	MeOH
	Other	Anticonvulsant	Carbamazepine	MeOH
			Oxcarbazepine	MeOH
		Antidepressant	Venlafaxine	MeOH
			Desmethylvenlafaxine	Acidic H_2O/MeOH (1/4)
		Antihistamine	Diphenhydramine	MeOH
			Fexofenadine	MeOH
		Antihypertensive	Amlodipine	MeOH

Class	Sub-class	Use	Compound	Stock solution solvent (*v/v*)
		Inhibitor of Angiotensin Converting Enzyme	Ramipril	MeOH
		Lipid-lowering agent	Atorvastatin	MeOH
		Synthetic thyroid hormone	Levothyroxine	DMSO
Artificial sweetener			Sucralose	MeOH
Personal care products		Insect repellent	N,N-Diethyl-3-methylbenzamide (DEET)	MeOH
		UV filter	Oxybenzone	MeOH
			Sulisobenzone	MeOH
Additives		Anticorrosive	5-Methyl-1H-benzotriazole	MeOH
		Plasticizer, flame retardant	Bisphenol A (BPA)	MeOH
			Tris(2-butoxyethyl) phosphate (TBEP)	MeOH
			Tributyl phosphate (TBP)	MeOH
			Tris(2-chloroethyl) phosphate (TCEP)	MeOH
			Tris(dichloroisopropyl) phosphate (TDCPP)	MeOH
			TPP (Triphenyl phosphate)	MeOH
Pesticides	Herbicide	Herbicide	2,4-Dichlorophenoxyacetic acid (2,4-D)	MeOH
			Atrazine	MeOH
			Bentazon	MeOH
			Bromoxynil	MeOH
			Dimethenamid	MeOH
			2-Methyl-4-chlorophenoxyacetic acid (MCPA)	MeOH
			Metolachlor	MeOH

Class	Sub-class	Use	Compound	Stock solution solvent (*v/v*)
			Pendimethalin	MeOH
			Triallate	MeOH
		Herbicide metabolite (atrazine)	Deethylatrazine	MeOH
	Insecticide	Insecticide	Chlorpyrifos	MeOH
			Clothianidin	MeOH
			Diazinon	MeOH
			Imidacloprid	MeOH
			Thiamethoxam	MeOH
		Insecticide metabolite (chlorpyrifos)	Chlorpyrifos oxon	MeOH

Tableau 8. Chromatographic gradients for the positive and negative ionization modes.

Positive mode			Negative mode		
Time (min)	A (%)	B (%)	Time (min)	A (%)	B (%)
0	98	2	0	85	15
1	98	2	1.2	64	36
11.4	12	88	2	46	54
11.8	0	100	2.6	40	60
14.2	0	100	3.4	0	100
15	98	2	5.8	0	100
17.4	98	2	6.2	85	15
			8.6	85	15

Tableau 9. MRM transitions for each contaminant and deuterated standard on the Xevo TQ (1) and TQ-S micro (2).

Analyte	Mass spectrometer	ESI polarity	Quantification (m/z)	Qualification (m/z)
1,7-Dimethylxanthine (Paraxanthine)	(1)	+	181.1 → 124.0	181.1 → 55.0
	(2)	+	181.0 → 124.0	181.0 → 55.0
2,4-Dichlorophenoxyacetic acid (2,4-D)	(1)	-	219.0 → 160.9	219.0 → 125.0
	(2)	-	218.9 → 160.9	218.9 → 124.9
5-Methyl-1H-benzotriazole	(1)	+	134.0 → 79.0	134.0 → 77.0
	(2)	+	134.0 → 77.0	134.0 → 79.0
Acetaminophen	(1)	+	152.1 → 110.1	152.1 → 93.0
	(2)	+	152.0 → 109.9	152.0 → 92.8
Acetaminophen-d3	(1)	+	155.0 → 111.0	ND
	(2)	+	155.0 → 111.0	ND
Amlodipine	(1)	+	409.3 → 238.0	409.3 → 294.0
	(2)	+	409.1 → 238.0	409.1 → 294.1
Anhydroerythromycin A	(1)	+	716.6 → 158.0	716.6 → 558.0
	(2)	+	716.4 → 158.1	716.4 → 116.0
Atorvastatin	(1)	+	559.2 → 440.0	559.2 → 250.0
	(2)	+	559.1 → 440.2	559.1 → 250.1
Atrazine	(1)	+	216.1 → 174.0	216.1 → 96.0
	(2)	+	216.1 → 174.1	216.1 → 96.0
Atrazine-d5	(1)	+	221.1 → 179.1	ND
	(2)	+	221.1 → 179.1	ND
Azithromycin	(1)	+	375.3 → 158.0	749.3 → 591.1
	(2)	+	375.3 → 158.0	749.3 → 83.1
Bentazon	(1)	-	239.1 → 132.0	239.1 → 197.1
	(2)	-	239.0 → 132.1	239.0 → 196.9
Bisphenol A (BPA)	(2)	-	227.1 → 212.0	227.1 → 133.0
Bromoxynil	(1)	-	275.8 → 80.7	275.8 → 167.0
	(2)	-	273.8 → 78.8	273.8 → 166.8
Caffeine	(1)	+	195.0 → 138.0	195.0 → 110.0

Analyte	Mass spectrometer	ESI polarity	Quantification (m/z)	Qualification (m/z)
	(2)	+	195.1 → 138.0	195.1 → 110.0
Caffeine-d3	(1)	+	198.1 → 138.0	ND
	(2)	+	198.1 → 138.0	ND
Carbamazepine	(1)	+	237.0 → 194.1	237.0 → 179.0
	(2)	+	237.1 → 193.6	237.1 → 179.0
Ceftiofur	(1)	+	524.0 → 241.3	524.0 → 210.0
	(2)	+	524.1 → 241.0	524.1 → 210.0
Chlorpyrifos	(1)	+	349.9 → 197.8	349.9 → 124.9
	(2)	+	349.9 → 96.9	349.9 → 197.9
Chlorpyrifos oxon	(1)	+	333.9 → 277.9	333.9 → 198.0
	(2)	+	333.8 → 197.9	333.8 → 277.9
Chlortetracycline	(2)	+	479.0 → 444.0	479.0 → 154.0
Ciprofloxacin	(1)	+	332.2 → 314.1	332.2 → 288.1
	(2)	+	332.0 → 288.1	332.0 → 314.1
Clofibric acid-d4	(1)	-	217.0 → 131.0	ND
	(2)	-	217.0 → 131.0	ND
Clothianidin	(1)	+	250.0 → 169.0	250.0 → 131.9
	(2)	+	250.0 → 168.9	250.0 → 131.9
Cotinine	(1)	+	177.1 → 80.0	177.1 → 98.0
	(2)	+	177.1 → 80.0	177.1 → 98.0
N,N-Diethyl-3-methylbenzamide (DEET)	(1)	+	192.1 → 119.0	192.1 → 91.0
	(2)	+	192.1 → 119.0	192.1 → 91.0
Deethylatrazine	(1)	+	188.1 → 145.9	188.1 → 78.9
	(2)	+	188.0 → 145.9	188.0 → 79.0
Desmethylvenlafaxine	(1)	+	264.1 → 58.0	264.1 → 246.1
	(2)	+	264.2 → 58.0	264.2 → 107.0
Diazinon	(1)	+	305.1 → 169.0	305.1 → 153.0
	(2)	+	305.0 → 169.1	305.0 → 153.1
Dimethenamid	(1)	+	276.0 → 244.3	276.0 → 168.0
	(2)	+	276.0 → 243.9	276.0 → 168.0

Analyte	Mass spectrometer	ESI polarity	Quantification (m/z)	Qualification (m/z)
Diphenhydramine	(1)	+	256.2 → 167.0	256.2 → 152.0
	(2)	+	256.1 → 167.0	256.1 → 152.0
Diphenhydramine-d4	(1)	+	261.1 → 172.2	ND
	(2)	+	261.2 → 172.1	ND
Fexofenadine	(1)	-	500.2 → 378.2	500.2 → 456.2
	(2)	-	500.2 → 456.4	500.2 → 378.2
Ibuprofen-d3	(1)	-	208.2 → 164.0	ND
	(2)	-	208.1 → 164.1	ND
Imidacloprid	(1)	+	256.0 → 209.0	256.0 → 175.1
	(2)	+	256.0 → 209.0	256.0 → 175.0
Levothyroxine	(1)	+	777.6 → 731.3	777.6 → 604.3
	(2)	+	777.5 → 731.6	777.5 → 323.7
Lincomycin	(1)	+	407.2 → 126.0	407.2 → 359.1
	(2)	+	407.1 → 126.1	407.1 → 359.1
2-Methyl-4-chlorophenoxyacetic acid	(1)	-	199.0 → 141.0	199.0 → 155.0
(MCPA)	(2)	-	198.9 → 140.9	198.9 → 154.9
Metolachlor	(1)	+	284.2 → 252.0	284.2 → 176.1
	(2)	+	284.0 → 252.1	284.0 → 176.0
Metolachlor-d6	(1)	+	290.2 → 258.2	ND
	(2)	+	290.1 → 258.1	ND
Naproxen	(1)	-	229.0 → 170.0	229.0 → 185.2
	(2)	-	229.0 → 170.0	229.0 → 185.1
Oxcarbazepine	(1)	+	253.1 → 236.0	253.1 → 208.0
	(2)	+	253.1 → 180.0	253.1 → 208.0
Oxybenzone	(1)	+	229.3 → 151.1	229.3 → 105.1
	(2)	+	229.0 → 151.0	229.0 → 104.9
Oxytetracycline	(2)	-	459.0 → 374.1	459.0 → 135.0
Pendimethalin	(1)	+	282.1 → 212.0	282.1 → 194.0
	(2)	+	282.1 → 212.0	282.1 → 194.1
Pendimethalin-d5	(1)	+	287.2 → 213.1	ND

Analyte	Mass spectrometer	ESI polarity	Quantification (m/z)	Qualification (m/z)
	(2)	+	287.1 → 212.9	ND
Ramipril	(1)	+	417.2 → 234.2	417.2 → 343.1
	(2)	+	417.2 → 234.1	417.2 → 117.0
Salicylic acid	(1)	-	137.1 → 93.0	137.1 → 65.0
	(2)	-	136.9 → 92.9	136.9 → 64.9
Sucralose	(1)	-	395.0 → 359.0	395.0 → 323.0
	(2)	-	394.9 → 359.0	394.9 → 143.1
Sulfamethazine	(1)	+	279.1 → 185.9	279.1 → 155.8
	(2)	+	279.1 → 186.0	279.1 → 92.0
Sulfamethoxazole	(1)	+	254.0 → 155.9	254.0 → 92.1
	(2)	+	254.0 → 92.1	254.0 → 156.0
Sulfamethoxazole-d4	(1)	+	258.1 → 160.0	ND
	(2)	+	258.1 → 96.0	ND
Sulfamethoxazole-d4	(1)	-	256.0 → 160.0	ND
	(2)	-	256.0 → 160.0	ND
Sulisobenzone	(1)	+	309.1 → 231.0	309.1 → 135.0
	(2)	+	309.0 → 231.0	309.0 → 134.9
Tris(2-butoxyethyl) phosphate (TBEP)	(1)	+	399.2 → 199.1	399.2 → 299.0
	(2)	+	399.2 → 199.1	399.2 → 101.0
Tributyl phosphate (TBP)	(1)	+	267.1 → 98.9	267.1 → 155.0
	(2)	+	267.1 → 98.9	267.1 → 155.0
Tris(2-chloroethyl) phosphate (TCEP)	(1)	+	284.9 → 98.9	284.9 → 161.0
	(2)	+	284.9 → 99.0	284.9 → 222.9
Tris(dichloroisopropyl) phosphate	(1)	+	431.4 → 99.0	431.4 → 209.0
(TDCPP)	(2)	+	431.3 → 99.0	431.3 → 209.0
Tetracycline	(2)	+	445.0 → 410.0	445.0 → 154.0
Thiamethoxam	(1)	+	292.0 → 211.0	292.0 → 181.0
	(2)	+	292.0 → 210.9	292.0 → 131.9
Triphenyl phosphate (TPP)	(1)	+	327.1 → 77.0	327.1 → 152.0
	(2)	+	327.0 → 77.0	327.0 → 152.0

Analyte	Mass spectrometer	ESI polarity	Quantification (m/z)	Qualification (m/z)
Triphenyl phosphate-d15	(1)	+	342.1 → 82.0	ND
	(2)	+	342.1 → 81.9	ND
Triallate	(1)	+	304.0 → 143.0	304.0 → 86.0
	(2)	+	304.0 → 86.0	304.0 → 142.9
Trimethoprim	(1)	+	291.2 → 230.1	291.2 → 123.1
	(2)	+	291.1 → 230.0	291.1 → 123.0
Venlafaxine	(1)	+	278.2 → 121.0	278.2 → 147.0
	(2)	+	278.2 → 121.0	278.2 → 91.0
Venlafaxine-d6	(1)	+	284.2 → 64.0	ND
	(2)	+	284.2 → 64.0	ND

ND: not determined

Tableau 10. Range of each figure of merit for all methodologies used for the 51 targeted analytes.

Analytes	Extraction recovery (%)	Precision (%)	LOD (ng/L)	LOQ (ng/L)	Blank 95th percentile (ng/L)	Deuterated IS
1,7-Dimethylxanthine (Paraxanthine)	87 - 93	2 - 26	0.3-30	1-100	1.2	Caffeine-D3
2,4-Dichlorophenoxyacetic acid (2,4-D)	96 - 109	13 - 23	0.3-173	1-571	0	Clofibric-D4 acid
5-Methyl-1H-benzotriazole	76 - 101	7 - 13	0.3-121	1-400	0	Sulfamethoxazole-D4
Acetaminophen	48 - 55	3 - 21	0.3-30	1-100	0	Acetaminophen-D3
Amlodipine	65 - 99	3 - 13	0.3-485	1-1600	0	Atrazine-D5
Anhydroerythromycin A	48 - 82	9 - 12	0.06-33	0.2-109	0	Atrazine-D5
Atorvastatin	19 - 33	24 - 28	0.61-586	2-1933	0	Metolachlor-D6
Atrazine	97 - 106	2 - 11	0.3-30	1-100	0	Atrazine-D5
Azithromycin	12 - 86	5 - 20	0.3-485	1-1600	23.1	Venlafaxine-D6
Bentazon	82 - 98	3 - 28	0.3-170	1-562	0	Sulfamethoxazole-D4
Bisphenol A (BPA)	110	4	0.06-121	0.2-400	78.5	Ibuprofen-D3

Analytes	Extraction recovery (%)	Precision (%)	LOD (ng/L)	LOQ (ng/L)	Blank 95th percentile (ng/L)	Deuterated IS
Bromoxynil	97 - 112	7 - 26	0.3-163	1-539	0	Sulfamethoxazole-D4
Caffeine	97 - 103	5 - 8	0.3-30	1-100	7.4	Caffeine-D3
Carbamazepine	94 - 98	3 - 20	0.06-121	0.2-400	2.0	Atrazine-D5
Ceftiofur	100 - 116	3 - 8	0.06-33	0.2-109	0	Venlafaxine-D6
Chlorpyrifos	21 - 35	9 - 27	0.61-1198	2-3955	0	TPP-D15
Chlorpyrifos oxon	94 - 101	9 - 30	0.3-165	1-546	0	Metolachlor-D6
Chlortetracycline	96	8	6.06-121	20-400	0	Sulfamethoxazole-D4
Ciprofloxacin	83 - 144	2 - 25	0.61-533	2-1760	26.3	Sulfamethoxazole-D4
Clothianidin	94 - 100	5 - 15	0.61-121	2-400	0	Sulfamethoxazole-D4
Cotinine	93 - 104	7 - 15	0.06-10	0.2-34	0	Acetaminophen-D3
N,N-Diethyl-3-methylbenzamide (DEET)	95 - 106	0 - 14	0.3-121	1-400	900.2	Atrazine-D5
Deethylatrazine	96 - 104	2 - 14	0.3-121	1-400	0	Sulfamethoxazole-D4
Desmethylvenlafaxine	97 - 99	3 - 13	0.06-121	0.2-400	0	Sulfamethoxazole-D4
Diazinon	88 - 105	2 - 42	0.61-650	2-2146	0	TPP-D15
Dimethenamid	98 - 103	2 - 10	0.06-15	0.2-50	0	Metolachlor-D6
Diphenhydramine	96 - 97	1 - 10	0.3-6	1-20	0	Diphenhydramine-D4
Fexofenadine	36 - 98	8 - 17	0.61-485	2-1600	0	Clofibric-D4 acid
Imidacloprid	105 - 109	9 - 24	0.61-121	2-400	0	Sulfamethoxazole-D4
Levothyroxine	90 - 105	1 - 17	0.3-121	1-400	0	Metolachlor-D6
Lincomycin	62 - 77	5 - 17	0.3-6	1-20	0	Caffeine-D3
2-Methyl-4-chlorophenoxyacetic acid (MCPA)	96 - 115	5 - 23	0.3-146	1-482	0	Clofibric-D4 acid
Metolachlor	95 - 105	2 - 15	0.06-30	0.2-100	0	Metolachlor-D6
Naproxen	104 - 107	8 - 41	0.61-995	2-3282	0	Clofibric-D4 acid
Oxcarbazepine	96 - 100	6 - 14	0.3-121	1-400	0	Atrazine-D5
Oxybenzone	85 - 106	7 - 73	0.06-704	0.2-2324	314.5	Metolachlor-D6
Oxytetracycline	100	1	6.06-121	20-400	0	Sulfamethoxazole-D4

Analytes	Extraction recovery (%)	Precision (%)	LOD (ng/L)	LOQ (ng/L)	Blank 95th percentile (ng/L)	Deuterated IS
Pendimethalin	21 - 37	9 - 23	0.61-74	2-245	0	Pendimethalin-D5
Ramipril	95 - 98	2 - 10	0.06-6	0.2-20	0	Atrazine-D5
Salicylic acid	72 - 105	17 - 23	0.06-144	0.2-476	550.4	Sulfamethoxazole-D4
Sucralose	93 - 96	15 - 36	0.01-485	0.02-1600	0	Sulfamethoxazole-D4
Sulfamethazine	93 - 101	2 - 12	0.01-121	0.02-400	0	Caffeine-D3
Sulfamethoxazole	95 - 103	3 - 11	0.06-30	0.2-100	0	Sulfamethoxazole-D4
Sulisobenzone	97 - 102	2 - 10	0.61-121	2-400	0	Venlafaxine-D6
Tris(2-butoxyethyl) phosphate (TBEP)	72 - 106	2 - 15	0.3-178	1-589	496.6	TPP-D15
Tributyl phosphate (TBP)	84 - 100	8 - 44	0.61-121	2-400	137.1	TPP-D15
Tris(2-chloroethyl) phosphate (TCEP)	85 - 97	0 - 12	0.3-30	1-100	293.0	Atrazine-D5
Tris(dichloroisopropyl) phosphate (TDCPP)	95 - 112	1 - 23	0.61-61	2-200	14.6	TPP-D15
Tetracycline	100	2	6.06-121	20-400	0	Caffeine-D3
Thiamethoxam	92 - 101	1 - 21	0.61-121	2-400	0	Caffeine-D3
Triphenyl phosphate (TPP)	62 - 96	2 - 34	0.3-61	1-200	323.4	TPP-D15
Triallate	55 - 99	3 - 31	0.06-755	0.2-2490	0	TPP-D15
Trimethoprim	95 - 105	1 - 9	0.3-121	1-400	2.0	Caffeine-D3
Venlafaxine	96 - 97	2 - 7	0.3-30	1-100	0	Venlafaxine-D6

Tableau 11. Detailed figures of merit are given for 4 validation methods: (1) Manual SPE followed by analysis on the Xevo TQ mass spectrometer before July 2018, (2) Manual SPE followed by analysis on

the Xevo TQ mass spectrometer from July 2018, (3) Automatic SPE with analysis on the Xevo TQ mass spectrometer, (4) Automatic SPE with analysis on the Xevo TQ-S micro mass spectrometer.

Analyte	Method	SPE recovery (%)	Precision (%)
1,7-Dimethylxanthine	(1), (2)	93%	7%
(Paraxanthine)	(3)	87%	26%
	(4)	93%	2%
2,4-Dichlorophenoxyacetic acid	(1), (2)	109%	14%
(2,4-D)	(3)	96%	23%
	(4)	98%	13%
5-Methyl-1H-benzotriazole	(1), (2)	101%	7%
	(3)	76%	12%
	(4)	100%	13%
Acetaminophen	(1)	91%	21%
	(2)	48%	21%
	(3)	53%	17%
	(4)	55%	3%
Amlodipine	(1), (2)	68%	11%
	(3)	65%	13%
	(4)	99%	3%
Anhydroerythromycin A	(1), (2)	82%	12%
	(3)	48%	9%
	(4)	48%	9%
Atorvastatin	(1), (2)	33%	28%
	(3)	32%	24%
	(4)	19%	28%
Atrazine	(1), (2)	97%	6%
	(3)	106%	11%
	(4)	102%	2%
Azithromycin	(1)	36%	10%
	(2)	86%	10%
	(3)	12%	20%

Analyte	Method	SPE recovery (%)	Precision (%)
	(4)	44%	5%
Bentazon	(1), (2)	96%	6%
	(3)	82%	28%
	(4)	98%	3%
Bisphenol A (BPA)	(4)	110%	4%
Bromoxynil	(1), (2)	101%	26%
	(3)	112%	19%
	(4)	97%	7%
Caffeine	(1), (2)	100%	6%
	(3)	103%	8%
	(4)	97%	5%
Carbamazepine	(1), (2)	98%	3%
	(3)	97%	13%
	(4)	94%	20%
Ceftiofur	(1), (2)	105%	3%
	(3)	116%	8%
	(4)	100%	4%
Chlorpyrifos	(1), (2)	35%	27%
	(3)	23%	24%
	(4)	21%	9%
Chlorpyrifos oxon	(1), (2)	94%	9%
	(3)	101%	9%
	(4)	96%	30%
Chlortetracycline	(4)	96%	8%
Ciprofloxacin	(1), (2)	144%	25%
	(3)	105%	17%
	(4)	83%	2%
Clothianidin	(1), (2)	94%	ND
	(3)	94%	15%
	(4)	100%	5%
Cotinine	(1), (2)	97%	8%

Analyte	Method	SPE recovery (%)	Precision (%)
	(3)	93%	15%
	(4)	104%	7%
N,N-Diethyl-3-methylbenzamide (DEET)	(1), (2)	99%	6%
	(3)	95%	14%
	(4)	106%	0%
Deethylatrazine	(1), (2)	104%	3%
	(3)	96%	14%
	(4)	99%	2%
Desmethylvenlafaxine	(1), (2)	97%	3%
	(3)	99%	13%
	(4)	99%	7%
Diazinon	(1), (2)	102%	14%
	(3)	105%	42%
	(4)	88%	2%
Dimethenamid	(1), (2)	98%	3%
	(3)	103%	10%
	(4)	102%	2%
Diphenhydramine	(1), (2)	96%	1%
	(3)	97%	10%
	(4)	96%	4%
Fexofenadine	(1), (2)	95%	17%
	(3)	36%	15%
	(4)	98%	8%
Imidacloprid	(3)	109%	9%
	(4)	105%	24%
Levothyroxine	(1), (2)	99%	17%
	(3)	90%	15%
	(4)	105%	1%
Lincomycin	(1), (2)	77%	5%
	(3)	74%	10%

Analyte	Method	SPE recovery (%)	Precision (%)
	(4)	62%	17%
2-Methyl-4-chlorophenoxyacetic acid (MCPA)	(1), (2)	115%	18%
	(3)	96%	23%
	(4)	98%	5%
Metolachlor	(1), (2)	97%	2%
	(3)	105%	15%
	(4)	95%	6%
Naproxen	(1), (2)	105%	41%
	(3)	104%	8%
	(4)	107%	40%
Oxcarbazepine	(1), (2)	98%	6%
	(3)	100%	14%
	(4)	96%	14%
Oxybenzone	(1), (2)	85%	73%
	(3)	93%	7%
	(4)	106%	16%
Oxytetracycline	(4)	100%	1%
Pendimethalin	(1), (2)	37%	23%
	(3)	34%	22%
	(4)	21%	9%
Ramipril	(1), (2)	98%	2%
	(3)	96%	10%
	(4)	95%	7%
Salicylic acid	(1), (2)	80%	17%
	(3)	105%	23%
	(4)	72%	23%
Sucralose	(1), (2)	95%	ND
	(3)	93%	36%
	(4)	96%	15%
Sulfamethazine	(1), (2)	97%	9%
	(3)	93%	12%

Analyte	Method	SPE recovery (%)	Precision (%)
	(4)	101%	2%
Sulfamethoxazole	(1), (2)	99%	3%
	(3)	103%	11%
	(4)	95%	3%
Sulisobenzone	(1), (2)	102%	3%
	(3)	97%	10%
	(4)	100%	2%
Tris(2-butoxyethyl) phosphate	(1), (2)	74%	6%
(TBEP)	(3)	72%	15%
	(4)	106%	2%
Tributyl phosphate (TBP)	(1), (2)	98%	19%
	(3)	84%	44%
	(4)	100%	8%
Tris(2-chloroethyl) phosphate	(1), (2)	97%	12%
(TCEP)	(3)	85%	11%
	(4)	93%	0%
Tris(dichloroisopropyl) phosphate	(1), (2)	95%	23%
(TDCPP)	(3)	112%	7%
	(4)	103%	1%
Tetracycline	(4)	100%	2%
Thiamethoxam	(3)	101%	21%
	(4)	92%	1%
Triphenyl phosphate (TPP)	(1), (2)	76%	2%
	(3)	62%	34%
	(4)	96%	2%
Triallate	(1), (2)	55%	31%
	(3)	61%	11%
	(4)	99%	3%
Trimethoprim	(1), (2)	95%	7%
	(3)	105%	9%
	(4)	98%	1%

Analyte	Method	SPE recovery (%)	Precision (%)
Venlafaxine	(1), (2)	97%	2%
	(3)	97%	7%
	(4)	96%	5%

Tableau 12. Concentrations above quantification limits of the target TrOCs in the sampled lakes. En raison du grand nombre de données, ce tableau est uniquement disponible en ligne au lien suivant : https://doi.org/10.1016/j.envpol.2024.123764.

Tableau 13. Criteria for the protection of aquatic life in freshwater regarding chronical effects in Quebec and for EU drinking water.

Analytes	Quebec criteria (ng/L)	EU criteria (ng/L)
1,7-Dimethylxanthine (Paraxanthine)		
2,4-Dichlorophenoxyacetic acid (2,4-D)	4000	100
5-Methyl-1H-benzotriazole		
Acetaminophen		
Amlodipine		
Anhydroerythromycin A		
Atorvastatin		
Atrazine	1800	100
Azithromycin		
Bentazon	510000	100
Bisphenol A (BPA)		2500
Bromoxynil	5000	100
Caffeine		
Carbamazepine	10000	
Ceftiofur		
Chlorpyrifos	2	100
Chlorpyrifos oxon		100
Chlortetracycline		
Ciprofloxacin		

Analytes	Quebec criteria (ng/L)	EU criteria (ng/L)
Clothianidin	8.3	100
Cotinine		
N,N-Diethyl-3-methylbenzamide (DEET)		
Deethylatrazine		100
Desmethylvenlafaxine		
Diazinon	4	100
Dimethenamid	5600	100
Diphenhydramine		
Fexofenadine		
Imidacloprid	8.3	100
Levothyroxine		
Lincomycin		
2-Methyl-4-chlorophenoxyacetic acid (MCPA)	2600	100
Metolachlor	7800	100
Naproxen	96000	
Oxcarbazepine		
Oxybenzone		
Oxytetracycline		
Pendimethalin		100
Ramipril		
Salicylic acid		
Sucralose		
Sulfamethazine		
Sulfamethoxazole		
Sulisobenzone		
Tris(2-butoxyethyl) phosphate (TBEP)		
Tributyl phosphate (TBP)		
Tris(2-chloroethyl) phosphate (TCEP)		
Tris(dichloroisopropyl) phosphate (TDCPP)		
Tetracycline		
Thiamethoxam	8.3	100

Analytes	Quebec criteria (ng/L)	EU criteria (ng/L)
Triphenyl phosphate (TPP)	4000	
Triallate	240	100
Trimethoprim		
Venlafaxine		

Tableau 14. PNEC and maximum rq for individual compounds for the 3 test species.

	P. promelas			*D. magna*			*T. pyriformis*		
Analytes	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*
1,7-Dimethylxanthine (Paraxanthine)	TEST	1851710	0.00	TEST	416690	0.00	NA	NA	NA
2,4-Dichlorophenoxyacetic acid (2,4-D)	Experimental	133000	0.00	Experimental	10000	0.06	TEST	24560	0.02
5-Methyl-1H-benzotriazole	Experimental	22000	0.03	Experimental	51600	0.01	NA	NA	NA
Acetaminophen	Experimental	813760	0.00	Experimental	4680	0.03	Experimental	998840	0.00
Amlodipine	TEST	730	0.05	TEST	3600	0.01	TEST	45730	0.00
Anhydroerythromycin A	NA	NA	NA	TEST	11190	0.00	NA	NA	NA
Atorvastatin	TEST	2	45.00	TEST	620	0.14	NA	NA	NA
Atrazine	Experimental	14990	0.03	Experimental	6900	0.07	Experimental	96000	0.00
Azithromycin	NA	NA	NA	Experimental	120100	0.00	NA	NA	NA
Bentazon	NA	NA	NA	TEST	43720	0.01	NA	NA	NA
Bisphenol A (BPA)	Experimental	4200	0.00	Experimental	7300	0.00	TEST	5300	0.00
Bromoxynil	Experimental	11500	0.01	Experimental	19220	0.00	Experimental	19160	0.00
Caffeine	Experimental	151000	0.00	Experimental	177490	0.00	NA	NA	NA
Carbamazepine	NA	NA	NA	Experimental	13790	0.01	NA	NA	NA
Ceftiofur	NA	NA	NA	NA	NA	NA	NA	NA	NA
Chlorpyrifos	Experimental	120	0.00	Experimental	0	0.00	NA	NA	NA
Chlorpyrifos oxon	TEST	150	0.00	TEST	24	0.00	TEST	52520	0.00
Chlortetracycline	TEST	320	0.00	Experimental	111200	0.00	NA	NA	NA
Ciprofloxacin	TEST	190	0.97	Experimental	1100	0.17	NA	NA	NA

	P. promelas			*D. magna*			*T. pyriformis*		
Analytes	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*
Clothianidin	NA	NA	NA	Experimental	11430	0.02	NA	NA	NA
Cotinine	TEST	199730	0.00	TEST	41950	0.00	TEST	413160	0.00
N,N-Diethyl-3-methylbenzamide (DEET)	Experimental	110080	0.01	Experimental	75000	0.02	TEST	64720	0.02
Deethylatrazine	TEST	105920	0.00	Experimental	35600	0.01	NA	NA	NA
Desmethylvenlafaxine	TEST	7540	0.02	TEST	6580	0.03	TEST	13970	0.01
Diazinon	Experimental	3700	0.00	Experimental	1	0.00	NA	NA	NA
Dimethenamid	NA	NA	NA	Experimental	16000	0.01	TEST	9960	0.02
Diphenhydramine	TEST	11340	0.00	TEST	1350	0.00	TEST	12360	0.00
Fexofenadine	TEST	59	0.00	TEST	4500	0.00	TEST	270	0.00
Imidacloprid	NA	NA	NA	Experimental	6029	0.00	NA	NA	NA
Levothyroxine	TEST	3	7.52	NA	NA	NA	NA	NA	NA
Lincomycin	TEST	10780	0.00	TEST	771870	0.00	NA	NA	NA
2-Methyl-4-chlorophenoxyacetic acid (MCPA)	TEST	32260	0.02	Experimental	180050	0.00	TEST	45210	0.01
Metolachlor	Experimental	8000	0.05	Experimental	4250	0.10	TEST	9450	0.05
Naproxen	TEST	4520	0.00	Experimental	37000	0.00	TEST	24990	0.00
Oxcarbazepine	TEST	9530	0.00	TEST	5470	0.00	NA	NA	NA
Oxybenzone	TEST	6820	0.07	TEST	4570	0.10	Experimental	8680	0.05
Oxytetracycline	TEST	1120	0.00	Experimental	621200	0.00	NA	NA	NA
Pendimethalin	TEST	190	0.52	Experimental	280	0.35	TEST	880	0.11

	P. promelas			*D. magna*			*T. pyriformis*		
Analytes	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*	Data source	PNEC (ng/L)	Max *rq*
Ramipril	TEST	410	0.00	TEST	24660	0.00	TEST	5260	0.00
Salicylic acid	TEST	98340	0.02	Experimental	111700	0.02	Experimental	446980	0.00
Sucralose	TEST	551280	0.00	TEST	1007670	0.00	NA	NA	NA
Sulfamethazine	NA	NA	NA	Experimental	31400	0.00	NA	NA	NA
Sulfamethoxazole	NA	NA	NA	Experimental	96700	0.00	NA	NA	NA
Sulisobenzone	TEST	2180	0.00	NA	NA	NA	NA	NA	NA
Tris(2-butoxyethyl) phosphate (TBEP)	Experimental	11200	0.13	TEST	95	15.40	NA	NA	NA
Tributyl phosphate (TBP)	Experimental	1000	0.20	Experimental	1170	0.17	TEST	197640	0.00
Tris(2-chloroethyl) phosphate (TCEP)	TEST	17310	0.00	TEST	22	0.00	TEST	250710	0.00
Tris(dichloroisopropyl) phosphate (TDCPP)	TEST	550	0.72	TEST	8	48.97	NA	NA	NA
Tetracycline	TEST	900	0.00	Experimental	170	0.00	NA	NA	NA
Thiamethoxam	NA	NA	NA	Experimental	126000	0.00	NA	NA	NA
Triphenyl phosphate (TPP)	Experimental	660	1.26	Experimental	90	9.21	Experimental	5050	0.16
Triallate	NA	NA	NA	Experimental	57	0.00	NA	NA	NA
Trimethoprim	TEST	10680	0.02	Experimental	92000	0.00	TEST	102130	0.00
Venlafaxine	TEST	6310	0.02	TEST	8180	0.01	TEST	8980	0.01

Tableau 15. Calculated *rq* for individual compounds in each sampled lake for the fish species *Pimephales promelas*.

En raison du grand nombre de données, ce tableau est uniquement disponible en ligne au lien suivant : https://doi.org/10.1016/j.envpol.2024.123764.

Tableau 16. Calculated *rq* for individual compounds in each sampled lake for the crustacean species *Daphnia magna*.

En raison du grand nombre de données, ce tableau est uniquement disponible en ligne au lien suivant : https://doi.org/10.1016/j.envpol.2024.123764.

Tableau 17. Calculated *rq* for individual compounds in each sampled lake for the ciliate species *Tetrahymena pyriformis*.

En raison du grand nombre de données, ce tableau est uniquement disponible en ligne au lien suivant : https://doi.org/10.1016/j.envpol.2024.123764.

Tableau 18. Potential for developmental toxicity and mutagenicity of each compound.

Analytes	Developmental toxicity	Mutagenicity
1,7-Dimethylxanthine (Paraxanthine)	Toxicant	Negative
2,4-Dichlorophenoxyacetic acid (2,4-D)	Toxicant	Negative
5-Methyl-1H-benzotriazole	Non-toxicant	Negative
Acetaminophen	Non-toxicant	Negative
Amlodipine	Toxicant	Negative
Anhydroerythromycin A	NA	Negative
Atorvastatin	Toxicant	Negative
Atrazine	Toxicant	Negative
Azithromycin	NA	Negative
Bentazon	Toxicant	Negative
Bisphenol A (BPA)	Toxicant	Negative
Bromoxynil	NA	Negative

Analytes	Developmental toxicity	Mutagenicity
Caffeine	Non-toxicant	Negative
Carbamazepine	Toxicant	Negative
Ceftiofur	Non-toxicant	NA
Chlorpyrifos	Toxicant	Negative
Chlorpyrifos oxon	Toxicant	Negative
Chlortetracycline	Toxicant	Positive
Ciprofloxacin	Toxicant	Positive
Clothianidin	Toxicant	Positive
Cotinine	Toxicant	Negative
N,N-Diethyl-3-methylbenzamide (DEET)	Toxicant	Negative
Deethylatrazine	Toxicant	Negative
Desmethylvenlafaxine	Toxicant	Negative
Diazinon	Toxicant	Negative
Dimethenamid	Toxicant	Negative
Diphenhydramine	Non-toxicant	Negative
Fexofenadine	Toxicant	Negative
Imidacloprid	Toxicant	Positive
Levothyroxine	Toxicant	NA
Lincomycin	Non-toxicant	Negative
2-Methyl-4-chlorophenoxyacetic acid (MCPA)	Toxicant	Negative
Metolachlor	Toxicant	Negative
Naproxen	Toxicant	Negative
Oxcarbazepine	Toxicant	Negative
Oxybenzone	Toxicant	Negative
Oxytetracycline	Toxicant	Positive
Pendimethalin	Toxicant	Negative
Ramipril	Toxicant	Negative
Salicylic acid	Non-toxicant	Negative
Sucralose	Toxicant	Negative
Sulfamethazine	Toxicant	Negative

Analytes	Developmental toxicity	Mutagenicity
Sulfamethoxazole	Toxicant	Negative
Sulisobenzone	Toxicant	Negative
Tris(2-butoxyethyl) phosphate (TBEP)	Toxicant	Negative
Tributyl phosphate (TBP)	Non-toxicant	Negative
Tris(2-chloroethyl) phosphate (TCEP)	Non-toxicant	Negative
Tris(dichloroisopropyl) phosphate (TDCPP)	Toxicant	Positive
Tetracycline	Toxicant	Positive
Thiamethoxam	Toxicant	Positive
Triphenyl phosphate (TPP)	Toxicant	Negative
Triallate	Non-toxicant	Positive
Trimethoprim	Toxicant	Negative
Venlafaxine	Toxicant	Negative

ANNEXE 2. INFORMATIONS SUPPLÉMENTAIRES DU CHAPITRE 3

Tableau 19. General information on the lakes included in the study.

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
04-572	Nataiinlaii Gwint'ii Van, NT	67.332779	-134.844522	0.5	moderate
04-577	Unnamed lake, NT	68.344096	-133.705674	0.1	high
04-579	Campbell Lake, NT	68.232838	-133.44037	35.7	low
04-580	Unnamed lake, NT	67.46993	-134.72108	0.7	moderate
04-583	Unnamed lake, NT	67.442962	-134.511877	0.3	low
04-609	Heart Lake, NT	60.836026	-116.656975	2.5	low
04-612	Unnamed lake, NT	60.721344	-114.996196	0.7	moderate
04-613	Unnamed lake, NT	60.813148	-114.589631	0.1	low
04-614	Unnamed lake, NT	60.53671	-114.568217	0.2	low
04-615	Pine Lake, AB	59.534902	-112.216745	3.0	low
04-616	Unnamed lake, AB	59.944209	-111.853586	0.4	low
04-617	Unnamed lake, NT	60.596644	-116.262865	0.3	low
04-618	Chan Lake, NT	61.890891	-116.542274	0.8	low
04-619	Unnamed lake, NT	62.556145	-116.433386	0.1	low
04-623	Parker Lake, BC	58.822968	-122.901342	0.6	low
04-624	Unnamed lake, BC	58.623863	-122.693773	0.2	low
06-083	Lac Saint-Antoine, QC	47.534528	-70.232947	0.3	moderate
06-085	Lac Nairne, QC	47.685661	-70.349677	2.5	moderate
06-102	Lac Froget, QC	50.732795	-71.771475	3.1	high
06-103	Lac à la Croix, QC	48.395105	-71.773708	0.7	high
06-104	Lac Vouzier, QC	48.407729	-71.78678	0.1	high
06-126	Lac Paula, QC	48.993311	-74.029437	0.6	high
06-127	Lac Duminy, QC	48.987717	-74.06735	0.2	moderate
06-128	Lac des Seigneurs, QC	45.862265	-74.115675	0.2	high
06-129	Lac Winsch, QC	50.032073	-74.221815	1.3	low
06-130	Lac Marois, QC	45.850978	-74.131647	1.0	high

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
06-132	Lac Sainte-Marie, QC	45.958676	-74.266369	2.1	low
06-133	Lac Duhamel, QC	46.142222	-74.638943	0.5	low
06-136	Lac Ouellette, QC	46.719648	-75.437263	0.6	high
06-137	Lac des Îles, QC	46.450343	-75.551099	16.7	low
06-139	Lac Vert, QC	45.902448	-75.605782	0.3	high
06-158	Lac Merlin, QC	48.893947	-76.881395	0.2	moderate
06-174	Lac Wasi, ON	46.138734	-79.225947	6.9	moderate
06-200	Unnamed lake, ON	48.238266	-81.040785	0.1	high
06-217	Borden Lake, ON	47.836499	-83.286417	16.6	low
06-220	Bright Lake, ON	46.272242	-83.30755	12.5	moderate
06-221	Sandbar Lake, ON	47.835215	-83.348195	0.3	moderate
06-254	Little Rushy Pond, NL	48.949782	-55.693761	0.4	high
06-258	Hogans Pond, NL	47.583623	-52.852923	0.6	high
06-261	Big Otter Pond, NL	47.399963	-53.04748	0.7	moderate
06-274	Rabbit Lake, ON	49.787686	-94.463764	0.9	high
06-291	Camp Lake, MB	55.128342	-101.103371	0.5	high
06-293	Liz Lake, MB	55.487013	-98.050248	2.0	moderate
06-304	L226, ON	49.688665	-93.747517	0.2	low
06-315	Johnson's Lake, ON	49.711052	-83.684753	0.4	high
07-002	Larkins Pond, PE	46.421872	-62.428828	0.3	low
07-003	Lake Verde, PE	46.23885	-62.885457	0.2	high
07-005	Long Lake, NS	45.902727	-64.155071	1.0	low
07-006	Collins Lake, NB	46.110489	-64.150702	0.7	low
07-007	Blair Lake, NS	45.799248	-64.210287	0.5	high
07-008	Morice Pond, NB	45.931138	-64.368213	1.5	moderate
07-009	Black River Lake, NS	44.930654	-64.418764	7.5	low
07-010	Gaspereau Lake, NS	44.953072	-64.563372	22.0	low
07-013	Colwell Round Lake, NS	44.845846	-64.598487	0.1	low
07-015	Spectacle Lake, NS	44.259495	-64.610661	1.0	moderate
07-020	Unnamed lake, NB	47.58432	-64.955436	0.2	low

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
07-022	Lake Mulgrave, NS	44.496459	-65.471246	14.5	low
07-023	Goose Lake, NS	43.603816	-65.527065	0.6	low
07-024	Barrington Lake, NS	43.612856	-65.573869	1.6	low
07-026	Crocker Lake, NB	46.89332	-65.727509	0.8	low
07-029	Ritchie Lake, NB	45.415706	-65.96754	0.2	high
07-030	Lily Lake, NB	45.290432	-66.057534	0.1	high
07-031	Lac Innocent, NS	44.273871	-66.075345	0.1	high
07-033	Napadogan Lake, NB	46.416483	-66.943837	0.3	moderate
07-035	McKendrick Lake, NB	46.852777	-66.361613	0.8	low
07-037	Moose Lake, NB	45.448897	-66.47072	0.7	low
07-040	McDougall Lake, NB	45.324348	-66.774486	3.1	low
07-052	Davidson Lake, NB	45.93731	-67.158731	2.1	moderate
07-228	Fletchers Lake, NS	44.842279	-63.611864	1.0	high
07-229	First Lake, NS	44.770926	-63.661881	0.9	high
07-230	Fenerty Lake, NS	44.831734	-63.719825	0.7	moderate
07-231	Moore Lake, NS	44.961142	-63.757733	0.1	moderate
07-236	Governor Lake, NS	44.642918	-63.701964	0.4	high
07-237	Five Island Lake, NS	44.664418	-63.806279	1.4	moderate
07-243	Lake Charles, NS	44.722618	-63.551142	1.4	high
07-247	MacPherson Lake, NS	45.433201	-61.420529	0.9	moderate
07-248	Shepherd Lake, NS	45.522507	-61.560681	0.3	low
07-250	Lake Ainslie, NS	46.132703	-61.185324	58.8	low
07-251	Unnamed lake, NS	46.170166	-60.014699	0.2	moderate
08-097	Lac Saint-Augustin, QC	46.749625	-71.392292	0.7	high
08-118	Lac Saint-Paul, QC	46.304437	-72.476705	2.9	high
08-134	Loch Garry, ON	45.254158	-74.709611	3.9	low
08-135	Lac Georges, ON	45.605917	-74.97345	0.6	moderate
08-145	Cedar Lake, ON	44.418957	-76.399919	0.2	moderate
08-147	Mississippi Lake, ON	45.035782	-76.184422	25.2	low
08-151	Devil Lake, ON	44.577578	-76.440764	11.0	low

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
08-154	Lacs Healy, QC	45.844902	-76.653917	0.1	low
08-155	Varty Lake, ON	44.391856	-76.813362	6.3	moderate
08-160	Stoco Lake, ON	44.473281	-77.28962	5.8	low
08-164	Stevenson Lake, ON	44.274574	-77.910623	0.2	moderate
08-166	Rice Lake, ON	44.1779	-78.179116	99.7	low
08-168	Sturgeon Lake, ON	44.474286	-78.687323	46.4	moderate
08-175	Sunova Lake, ON	43.194682	-81.02096	0.3	high
08-177	Lake St. John, ON	44.687068	-79.325113	6.8	moderate
08-179	Wilcox Lake, ON	43.949049	-79.436034	0.6	high
08-180	Eversley Lake, ON	43.957772	-79.500823	0.1	high
08-182	Little Lake, ON	44.426424	-79.671388	2.4	high
08-184	Heart Lake, ON	43.740539	-79.795426	0.2	moderate
08-186	Lake Niapenco, ON	43.105298	-79.850926	1.9	high
08-187	Farden Lake, ON	44.276532	-80.690839	0.1	moderate
08-190	Pinehurst Lake, ON	43.269235	-80.390052	0.1	moderate
08-192	Green Lake, ON	43.839801	-80.008756	0.1	high
08-193	Dankert Lake, ON	44.205289	-81.052711	0.2	high
08-202	Boat Lake, ON	44.725929	-81.227067	5.6	moderate
08-205	Gillies Lake, ON	45.204954	-81.326861	2.2	high
08-206	Cyprus lake, ON	45.231341	-81.531897	0.8	low
08-208	Clam Lake, ON	44.072133	-81.413948	0.4	high
08-210	Unnamed lake, ON	43.575686	-81.661215	0.2	high
08-211	Sucker Lake, ON	45.723555	-81.878484	2.3	low
08-212	Pike Lake, ON	45.87402	-81.985603	2.3	low
08-214	Unnamed lake, ON	44.007233	-78.029579	0.1	moderate
08-218	Falls Lake, ON	45.898543	-83.106275	1.1	low
08-219	Lac Mccord, QC	45.693047	-76.46411	0.1	low
09-286	Nut Lake, SK	52.343345	-103.706775	18.0	low
09-292	Egg Lake, MB	54.37048	-101.457133	30.6	low
09-297	Devils Lake, MB	52.398538	-98.912922	0.5	low

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
09-369	Unnamed lake, AB	53.542469	-117.070949	0.1	moderate
09-370	Fickle Lake, AB	53.449373	-116.779894	3.6	low
09-375	Lac la Nonne, AB	53.939715	-114.321366	12.5	moderate
09-385	Jones Lake, AB	55.391549	-119.003383	1.3	moderate
09-392	Unnamed lake, AB	54.894823	-112.316588	0.2	moderate
09-393	Claude Lake, AB	54.793696	-111.909232	0.7	moderate
09-397	Upper Thérien Lake, AB	53.965983	-111.293303	8.1	high
09-409	Falcon Lake, MB	50.50351	-99.953734	0.3	high
09-411	Little Jackfish Lake, MB	50.47746	-100.074738	1.6	high
09-427	Poplar Ridge Lake, SK	53.92408	-107.702852	0.1	low
09-428	Green Lake, SK	54.09185	-107.6804	27.8	low
09-430	Unnamed lake, SK	54.993693	-108.344502	0.4	low
09-449	Unnamed lake, MB	51.439365	-101.42674	0.1	high
09-450	Meeting Lake, SK	53.188577	-107.657995	10.8	moderate
09-596	Pigeon Lake, AB	53.024412	-114.059639	94.8	low
09-602	Willow Lake, AB	56.462556	-111.162013	25.8	low
09-603	Kearl Lake, AB	57.292089	-111.238794	5.1	high
09-604	Saskatoon Lake, AB	55.217558	-119.091379	8.0	high
09-607	Kakut Lake, AB	55.628822	-118.528498	3.6	moderate
10-283	Bennet Lake, SK	49.771608	-102.458801	0.3	low
10-284	Unnamed lake, SK	49.939593	-102.451854	0.9	moderate
10-301	North Shoal Lake, MB	50.470346	-97.650318	31.9	low
10-346	Beaverdam Lake, AB	49.084553	-113.606695	0.7	low
10-349	Unnamed lake, AB	51.591071	-113.753331	0.3	high
10-352	Namaka Lake, AB	50.939508	-113.232916	6.4	moderate
10-355	Lac Pelletier, SK	49.986537	-107.934986	3.0	high
10-356	Fife Lake, SK	49.217733	-105.851938	29.4	moderate
10-357	Unnamed lake, SK	50.21254	-103.584538	0.5	high
10-358	Little Kenosee Lake, SK	49.83338	-102.332239	1.5	low
10-359	Alkali Lake, SK	49.813839	-102.397321	0.1	low

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
10-387	Driedmeat Lake, AB	52.85999	-112.74476	12.1	high
10-388	Boag Lake, AB	53.518523	-113.219575	0.9	moderate
10-389	Skaro Lake, AB	53.924854	-112.714336	0.4	high
10-398	Birch Lake, AB	53.316011	-111.589784	8.2	moderate
10-399	Thomas Lake, AB	53.110315	-111.699187	5.1	high
10-400	Macklin Lake, SK	52.320211	-109.95027	0.8	moderate
10-402	Miquelon Lakes, AB	53.256011	-112.912167	7.7	low
10-406	Red Deer Lake, AB	52.721431	-113.060046	21.1	moderate
10-412	Shoal Lake, MB	50.388326	-100.626474	4.8	high
10-416	Unnamed lake, SK	52.239307	-103.947363	0.6	moderate
10-433	Unnamed lake, SK	52.635863	-108.567479	0.2	high
10-435	Manitou Lake, SK	52.738966	-109.652625	81.2	moderate
10-438	Crookshanks Lake, SK	51.860856	-109.311232	0.3	high
10-444	Murray Lake, SK	53.037189	-108.278678	11.6	moderate
10-445	Schmidt Lake, SK	52.670482	-109.211384	0.5	moderate
10-452	Lytwyns Lake, MB	51.597074	-99.849842	0.3	low
10-453	Unnamed lake, SK	52.671989	-108.310133	0.5	low
10-597	Muir Lake, AB	53.623604	-113.95501	0.3	moderate
10-598	Astotin Lake, AB	53.678768	-112.853935	5.5	low
11-342	Whiteswan Lake, BC	50.144066	-115.474903	4.1	low
11-470	Dunalter Lake, BC	54.470753	-126.755752	0.2	moderate
11-478	Ferguson Lake, BC	54.037199	-122.84621	0.2	low
11-480	Hobson Lake, BC	53.580617	-124.73149	0.6	low
11-481	Fish Lake, BC	53.598731	-124.889241	0.4	low
11-482	Unnamed lake, BC	53.875399	-124.755299	0.1	low
11-483	Co-op Lake, BC	54.186152	-125.428674	0.3	low
11-484	Unnamed lake, BC	53.977642	-125.898396	0.3	high
11-486	Unnamed lake, BC	53.9938	-125.981445	0.2	moderate
11-487	Totem Pole Lake, BC	53.966067	-125.944652	0.3	moderate
11-488	Tatalaska Lake, BC	53.938458	-125.907128	1.6	high

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
11-489	Mollice Lake, BC	53.957671	-125.714231	1.9	moderate
11-491	Chief Lake, BC	54.120806	-123.004988	7.1	moderate
11-493	Unnamed lake, BC	54.008382	-123.135854	0.1	moderate
11-495	Williams Lake, BC	52.118825	-122.075003	6.8	low
11-499	Konni Lake, BC	51.473574	-123.887461	5.6	low
11-503	Clearwater Lake, BC	52.011725	-125.008155	2.1	low
11-508	Italia Lake, BC	51.83278	-120.388941	1.3	moderate
11-509	Latremouille Lake, BC	51.491893	-120.341789	0.8	low
11-541	Takatoot Lake, BC	55.120772	-125.196624	7.2	moderate
11-546	Unnamed lake, BC	54.168663	-124.208658	0.1	moderate
11-547	Naltesby Lake, BC	53.607447	-123.488697	8.2	low
11-556	Sepa Lake, BC	51.734097	-121.352237	0.2	high
11-557	Greeny Lake, BC	51.850947	-121.33891	0.8	low
11-589	Tudyah Lake, BC	55.084266	-123.038133	6.2	low
11-627	Huble Lake, BC	54.269426	-122.616376	0.2	low
11-628	Dominion Lake, BC	54.450825	-122.695605	0.7	low
11-629	Summit Lake, BC	54.277084	-122.678241	12.9	low
11-630	Rainbow Lake, BC	52.996317	-123.60365	0.7	moderate
11-633	Eight Mile Lake, BC	53.149595	-121.534574	0.1	moderate
12-460	Mill Lake, BC	49.044741	-122.310811	0.2	high
12-462	Deer Lake, BC	49.236235	-122.97178	0.3	high
12-466	Lillooet Lake, BC	50.215541	-122.490763	31.9	low
12-473	Kitseguecla Lake, BC	54.934739	-127.55412	0.7	low
12-504	Chehalis Lake, BC	49.443092	-122.020294	6.3	low
12-528	Clements Lake, BC	56.051194	-129.902444	0.2	low
12-529	Jigsaw Lake, BC	55.83382	-128.830233	0.7	low
12-530	Derrick Lake, BC	55.649296	-128.644809	0.5	low
12-531	Lava Lake, BC	55.047296	-128.993528	5.2	low
12-533	Unnamed lake, BC	55.445634	-129.353516	0.1	low
12-534	Unnamed lake, BC	55.446225	-129.334999	0.2	low

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
12-535	Unnamed lake, BC	55.4549	-129.36962	0.4	low
12-537	Taltzen Lake, BC	54.9301	-127.513355	0.1	low
12-636	Thetis Lake, BC	48.467719	-123.4691	0.2	moderate
12-637	Lake Weston, BC	48.784186	-123.425023	0.2	moderate
12-640	Buck Lake, BC	48.772668	-123.300605	0.1	high
12-641	Magic Lake, BC	48.763747	-123.288779	0.1	high
12-642	Langford Lake, BC	48.448187	-123.529102	0.6	high
12-644	Kemp Lake, BC	48.379522	-123.780515	0.2	moderate
12-646	Dougan Lake, BC	48.714643	-123.613474	0.1	high
12-648	Quamichan Lake, BC	48.799616	-123.662384	2.9	high
12-649	Fuller Lake, BC	48.908326	-123.720615	0.2	high
12-651	Holden Lake, BC	49.103249	-123.829263	0.4	moderate
12-653	Boomerang Lake, BC	49.17875	-124.155461	0.1	moderate
12-654	Kennedy Lake, BC	49.062864	-125.491128	65.3	low
12-655	Lowry Lake, BC	49.390048	-125.135344	0.4	low
12-656	Enos Lake, BC	49.28058	-124.156487	0.2	moderate
12-657	Green Lake, BC	49.230739	-124.060791	0.1	moderate
12-658	Mohun Lake, BC	50.11693	-125.499304	6.2	low
12-659	McCreight Lake, BC	50.301082	-125.642773	2.8	low
12-662	Victoria Lake, BC	50.371904	-127.387341	15.7	low
12-665	Zeballos Lake, BC	50.075821	-126.754403	2.0	low
12-670	Diver Lake, BC	49.203775	-124.014124	0.1	high
12-671	Brannen Lake, BC	49.214698	-124.055142	1.1	moderate
12-672	Long Lake, BC	49.210988	-124.017319	0.4	high
12-675	Horne Lake, BC	49.333971	-124.675007	8.1	low
12-676	Cranby Lake, BC	49.695238	-124.507497	0.4	moderate
12-677	Haslam Lake, BC	49.936063	-124.420312	11.6	low
13-514	Upper Gnat Lake, BC	58.21637	-129.837323	0.4	moderate
13-516	Good Hope Lake, BC	59.300477	-129.276001	1.7	low
13-517	Second Wye Lake, YT	60.062737	-128.680252	0.3	moderate

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
13-519	Watson Lake, YT	60.107692	-128.80679	13.4	low
13-521	Lower McDonald Lake, BC	59.703067	-133.60852	0.5	low
13-523	Bennett Lake, YT	60.118864	-134.842427	91.1	low
13-525	Dezadeash Lake, YT	60.479871	-137.010954	74.2	low
13-526	Pine Lake, YT	60.816837	-137.448355	5.8	low
13-560	Allan Lake, BC	58.430235	-130.001241	0.4	moderate
13-562	Unnamed lake, YT	60.080432	-128.745812	0.2	moderate
13-563	Unnamed lake, YT	61.455652	-129.745014	0.1	moderate
13-565	Jackfish Lake, YT	61.934997	-132.521865	1.1	low
13-566	Fish Eye Lake, YT	62.191856	-133.474391	0.4	moderate
13-568	Ethel Lake, YT	63.36165	-136.062415	43.3	low
13-569	Minto Lake, YT	63.685626	-136.161694	4.0	low
13-587	Unnamed lake, YT	60.450307	-134.265322	0.1	low
14-585	Two Moose Lake, YT	64.735027	-138.365273	0.1	high
17-038	Middle Peaked Mountain Lake, NB	46.736363	-66.519057	0.1	moderate
17-043	Trousers Lake, NB	47.010988	-66.970229	11.1	moderate
17-050	Lac des Indiens, QC	50.202718	-66.174349	0.1	high
17-054	Eightmile Lake, NB	47.695087	-67.644799	0.2	high
17-056	Lac Désiré, QC	48.674467	-67.734944	0.4	low
17-059	Lac de Saint-Damase, QC	48.653185	-67.809428	0.7	high
17-069	Lac Jerry, QC	47.429633	-68.785243	5.8	low
17-070	Lac Témiscouata, QC	47.686757	-68.848319	67.7	low
17-072	Grand lac Malobès, QC	48.269232	-68.860309	1.7	moderate
17-075	Lac du Marin-à-Gouin, QC	48.075148	-69.106521	0.2	moderate
17-076	Lac Dole, QC	47.645885	-68.944567	0.3	moderate
17-079	Lac Morin, QC	47.627411	-69.533598	6.0	low
17-081	Lac de l'Est, QC	47.186479	-69.561309	7.5	low
17-084	Lac du Portage, QC	45.935951	-70.270307	4.3	low
17-087	Lac Mailloux, QC	46.706288	-70.490235	0.2	moderate

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class
17-088	Lac Etchemin, QC	46.389586	-70.493319	2.5	moderate
17-090	Lac Volet, QC	46.137727	-70.809582	0.3	low
17-092	Lac Mégantic, QC	45.537896	-70.889698	28.3	moderate
17-093	Lac des Trois Milles, QC	45.687963	-70.921142	1.0	moderate
17-094	Lac Jolicoeur, QC	46.061894	-71.10254	0.2	high
17-098	Lac à la Truite, QC	46.084538	-71.500559	1.2	moderate
17-105	Petit lac Saint-François, QC	45.537697	-72.037776	0.8	high
17-107	Étang Burbank, QC	45.780093	-72.00684	0.5	high
17-109	Lac Magog, QC	45.305175	-72.041315	11.0	high
17-110	Lac Crystal, QC	45.030153	-72.076808	0.2	high
17-111	Lac Montjoie, QC	45.408466	-72.098912	3.4	low
17-114	Lac Brompton, QC	45.433218	-72.144938	11.2	low
17-117	Lac Memphrémagog, QC	45.026848	-72.246826	99.0	moderate
17-119	Lac Brome, QC	45.247947	-72.514469	15.1	moderate
17-123	Lac Bromont, QC	45.265997	-72.671014	0.5	moderate
18-324	Kalamalka Lake, BC	50.172799	-119.327369	25.4	high
18-335	Nicola Lake, BC	50.166411	-120.528153	24.9	low
18-336	Munro Lake, BC	49.715267	-119.923178	0.1	low
18-337	Skaha Lake, BC	49.410038	-119.585408	19.5	moderate
18-338	Green Lake, BC	49.302628	-119.57101	0.1	moderate
18-339	Vaseux Lake, BC	49.288771	-119.531568	2.8	high
18-505	Chapperon Lake, BC	50.202061	-120.055602	4.3	low
18-552	Stake Lake, BC	50.512281	-120.47799	0.2	high
18-553	Strachan Lake, BC	50.903978	-120.610563	0.1	high
18-593	Pennask Lake, BC	49.995289	-120.136936	9.4	low

Tableau 20. Detailed variables used in the statistical models.

	Dependent variables					Independent variables						Control variables					
Lake ID	Number of detections	Total TrOCs sum	Pesticides sum	PPCPs and additives sum	Antibiotics sum	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density	Area ratio	Lake depth	Residence time	Mean slope	Precipitation	Sampling date
lakeID	nb_overall	sum_overall	sum_pesticides	sum_parmaceuticals	sum_antibiotics	agriculture_frac	urban_frac	pop_density	wwtps_pres	hospitals_pres	livestock_density	area_ratio	lake_depth	res_time	mean_slope	precipitationSum7daysPrior	sampling_date
04-572	2	0	0	0	0	0.00	0.02	0	0	0	0	0.38	6	0	5.62	0.45	2019-07-31
04-577	2	0	0	0	0	0.00	0.25	5	0	0	0	0.25	6	973	3.72	1.23	2019-08-05
04-579	1	0	0	0	0	0.00	0.00	0	0	0	0	0.03	15	1177	2.09	1.61	2019-08-07
04-580	0	0	0	0	0	0.00	0.03	0	0	0	0	0.37	1	1965	0.73	0.24	2019-07-30
04-583	0	0	0	0	0	0.00	0.01	0	0	0	0	0.01	6	1620	1.92	0.41	2019-08-02
04-609	1	28	0	28	28	0.00	0.01	0	0	0	0	0.25	2	11893	0.58	1.99	2019-07-30
04-612	0	0	0	0	0	0.00	0.01	0	0	0	0	0.20	7	1825	1.50	0.71	2019-07-27

	Dependent variables					Independent variables						Control variables					
04-613	2	1	0	1	0	0.00	0.00	0	0	0	0	0.06	2	NA	0.69	0.75	2019-07-28
04-614	1	22	0	22	0	0.00	0.00	0	0	0	0	0.60	2	3204	0.07	0.85	2019-07-29
04-615	0	0	0	0	0	0.00	0.02	0	0	0	0	0.13	22	45171	1.00	3.34	2019-08-01
04-616	0	0	0	0	0	0.00	0.01	0	0	0	0	0.06	1	12678	0.30	1.95	2019-08-02
04-617	1	27	0	27	27	0.00	0.00	0	0	0	0	0.24	1	45500	0.44	1.16	2019-08-03
04-618	0	0	0	0	0	0.00	0.00	0	0	0	0	0.02	2	483	0.90	1.98	2019-08-07
04-619	0	0	0	0	0	0.00	0.01	0	0	0	0	0.01	1	32	1.17	1.98	2019-08-05
04-623	0	0	0	0	0	0.02	0.02	3	0	0	1	0.05	1	93	6.74	1.62	2019-08-12
04-624	0	0	0	0	0	0.00	0.02	0	0	0	0	0.47	2	1017	1.15	1.72	2019-08-13
06-083	7	105	11	94	0	0.02	0.00	6	0	0	23	0.04	4	84	11.75	2.06	2017-08-19
06-085	3	39	0	39	0	0.00	0.02	15	0	0	8	0.09	11	805	8.00	1.80	2017-08-18

	Dependent variables					Independent variables						Control variables					
06-102	8	135	96	40	0	0.00	0.00	0	0	0	0	0.15	13	465	7.68	7.89	2017-08-17
06-103	8	46	46	0	0	0.26	0.01	18	0	0	27	0.07	34	236	4.85	2.83	2017-08-22
06-104	7	39	10	28	3	0.57	0.19	199	0	0	24	0.18	37	32	4.14	2.62	2017-08-20
06-126	4	1	1	0	0	0.00	0.00	0	0	0	0	0.16	17	575	6.89	6.69	2017-08-25
06-127	7	62	34	27	0	0.00	0.00	0	0	0	0	0.02	2	35	3.91	5.42	2017-08-26
06-128	9	109	49	60	0	0.00	0.38	274	0	0	0	0.10	19	575	8.75	3.88	2017-07-14
06-129	5	628	0	628	0	0.00	0.01	0	0	0	0	0.12	33	169	5.37	6.65	2017-08-24
06-130	13	185	40	145	127	0.00	0.26	176	0	0	0	0.11	24	517	7.55	4.40	2017-07-13
06-132	9	69	24	45	0	0.00	0.05	29	1	0	0	0.02	16	222	17.05	2.77	2017-07-16
06-133	7	53	34	19	0	0.00	0.06	44	0	0	0	0.17	26	399	18.44	1.72	2017-07-17
06-136	3	7	5	1	0	0.11	0.04	5	0	0	19	0.15	13	393	4.75	3.11	2017-08-28

	Dependent variables					Independent variables						Control variables					
06-137	3	5	5	0	0	0.00	0.01	8	0	0	2	0.11	40	1723	8.28	3.05	2017-08-29
06-139	9	37	16	21	0	0.00	0.34	131	0	0	1	0.25	26	192	10.52	0.10	2017-08-30
06-158	2	9	9	0	0	0.00	0.00	0	0	0	0	0.01	6	27	5.95	2.46	2017-08-21
06-174	10	85	5	80	0	0.02	0.02	4	0	0	2	0.05	5	324	5.66	0.77	2017-09-02
06-200	2	21	21	0	0	0.00	0.00	0	0	0	0	0.10	3	589	6.02	3.21	2017-08-13
06-217	3	29	28	2	0	0.00	0.01	1	0	0	0	0.19	28	1245	3.44	5.09	2017-08-12
06-220	7	50	46	4	4	0.00	0.02	2	0	0	4	0.07	11	1295	6.03	4.27	2017-08-10
06-221	11	18	14	4	3	0.00	0.09	3	0	0	0	0.08	7	184	3.96	5.05	2017-08-11
06-254	6	302	0	302	0	0.00	0.17	66	0	0	0	0.27	3	1114	0.74	1.65	2018-08-08
06-258	5	21	0	21	0	0.00	0.38	150	0	0	2	0.29	12	362	5.85	0.76	2018-08-12
06-261	6	102	0	102	102	0.00	0.04	1	0	0	0	0.03	9	58	7.16	0.53	2018-08-13

	Dependent variables					Independent variables						Control variables					
06-274	1	27	0	27	27	0.00	0.36	376	0	0	0	0.34	9	12386	4.16	4.77	2018-07-05
06-291	5	69	0	69	69	0.00	0.01	0	0	0	0	0.00	3	66	4.33	6.71	2018-07-26
06-293	7	1670	0	1670	0	0.00	0.01	1	0	0	0	0.15	18	8255	3.46	4.73	2018-07-28
06-304	3	0	0	0	0	0.00	0.00	0	0	0	0	0.18	11	8702	11.85	0.17	2018-08-14
06-315	0	0	0	0	0	0.00	0.07	3	0	0	0	0.60	9	670	0.07	2.99	2018-08-28
07-002	3	0	0	0	0	0.06	0.01	4	0	0	2	0.01	4	17	2.60	0.03	2017-07-21
07-003	2	121	121	0	0	0.17	0.04	22	0	0	11	0.14	1	316	1.35	0.16	2017-07-20
07-005	3	1	0	1	0	0.00	0.01	1	0	0	0	0.05	2	251	0.22	0.92	2017-07-23
07-006	2	8	3	5	5	0.00	0.01	3	0	0	1	0.27	5	178	1.49	1.04	2017-07-24
07-007	18	89	23	66	8	0.01	0.34	88	0	0	5	0.06	5	145	2.20	0.80	2017-07-29
07-008	4	9	3	6	0	0.00	0.07	20	0	0	4	0.03	3	36	3.45	0.83	2017-07-28

	Dependent variables					Independent variables						Control variables					
07-009	2	0	0	0	0	0.00	0.01	2	0	0	0	0.02	12	262	4.61	2.61	2017-08-13
07-010	4	14	4	9	0	0.00	0.01	2	0	0	0	0.11	15	459	4.27	2.52	2017-08-11
07-013	1	0	0	0	0	0.00	0.00	1	0	0	0	0.08	2	121	5.04	2.49	2017-08-14
07-015	3	5	3	2	0	0.00	0.06	24	0	0	1	0.08	4	353	5.47	1.98	2017-08-18
07-020	4	10	0	10	0	0.00	0.02	21	0	0	0	0.01	3	11	2.71	0.62	2017-07-17
07-022	2	2	0	2	0	0.00	0.00	0	0	0	0	0.13	6	337	3.99	1.55	2017-08-09
07-023	5	9	2	8	0	0.00	0.01	1	0	0	0	0.02	1	35	4.13	4.62	2017-08-19
07-024	1	1	0	1	0	0.00	0.00	0	0	0	0	0.01	2	14	2.50	5.48	2017-08-20
07-026	1	4	0	4	0	0.00	0.00	3	0	0	0	0.23	2	403	0.73	0.39	2017-07-15
07-029	4	503	503	0	0	0.00	0.49	471	0	0	1	0.04	12	98	5.52	0.09	2017-08-05
07-030	8	29	13	16	0	0.00	0.31	184	0	0	0	0.15	8	NA	6.33	0.20	2017-08-04

	Dependent variables					Independent variables						Control variables					
07-031	7	767	0	767	0	0.00	0.15	41	0	0	12	0.32	3	716	4.17	4.47	2017-08-25
07-033	5	8	2	6	0	0.00	0.03	4	0	0	0	0.04	2	118	5.33	0.43	2017-08-02
07-035	6	6	6	0	0	0.00	0.02	0	0	0	0	0.10	6	291	6.45	1.79	2017-07-23
07-037	3	3	3	0	0	0.00	0.01	0	0	0	0	0.19	4	498	3.06	1.19	2017-08-06
07-040	6	65	3	61	0	0.00	0.01	0	0	0	0	0.03	4	76	5.21	0.72	2017-08-27
07-052	8	4	2	3	0	0.00	0.05	4	0	0	0	0.25	7	968	4.11	0.17	2017-08-30
07-228	9	28	0	28	0	0.00	0.25	166	1	0	0	0.01	9	326	5.65	1.50	2018-07-20
07-229	11	971	28	943	0	0.00	0.69	1498	0	0	0	0.22	23	702	5.46	1.72	2018-07-23
07-230	28	8162	526	7636	513	0.00	0.23	134	1	0	0	0.03	8	78	6.35	0.34	2018-07-15
07-231	22	5492	151	5341	88	0.00	0.03	13	0	0	0	0.14	2	368	5.95	0.41	2018-07-08
07-236	4	107	0	107	0	0.00	0.47	458	0	0	0	0.05	15	82	5.96	0.12	2018-07-16

	Dependent variables					Independent variables						Control variables					
07-237	1	311	0	311	0	0.00	0.19	76	0	0	0	0.07	9	85	4.20	1.76	2018-07-22
07-243	5	65	0	65	40	0.00	0.40	324	0	0	0	0.07	28	198	5.16	0.17	2018-07-14
07-247	4	0	0	0	0	0.00	0.07	3	0	0	0	0.21	21	180	6.71	0.97	2018-07-30
07-248	6	34	2	32	32	0.00	0.02	5	0	0	0	0.09	4	106	3.48	0.84	2018-07-29
07-250	1	0	0	0	0	0.00	0.01	1	0	0	1	0.19	16	961	9.01	1.44	2018-08-02
07-251	4	0	0	0	0	0.00	0.05	14	0	0	0	0.02	15	11	2.91	0.50	2018-08-03
08-097	11	154	77	76	0	0.05	0.58	860	0	0	8	0.12	6	331	2.59	4.94	2017-08-26
08-118	21	448	401	47	28	0.44	0.03	30	1	0	16	0.04	3	39	0.92	2.82	2017-08-20
08-134	7	22	20	2	0	0.00	0.01	9	0	0	3	0.24	4	565	1.56	1.83	2017-09-11
08-135	18	1430	1386	43	28	0.37	0.02	9	0	0	19	0.02	10	48	1.91	1.58	2017-09-12
08-145	8	179	74	105	0	0.00	0.08	67	0	0	12	0.17	14	717	4.91	2.46	2017-07-21

	Dependent variables					Independent variables						Control variables					
08-147	7	21	20	2	0	0.01	0.02	5	0	0	1	0.01	8	52	6.50	2.42	2017-09-09
08-151	6	47	29	18	0	0.00	0.02	5	0	0	1	0.06	41	1341	7.37	2.98	2017-07-16
08-154	7	14	14	0	0	0.00	0.00	2	0	0	0	0.25	21	NA	6.38	1.39	2017-09-03
08-155	5	63	54	10	0	0.04	0.04	17	0	0	8	0.28	2	749	1.38	1.93	2017-07-22
08-160	7	77	61	16	0	0.01	0.02	5	1	0	3	0.00	14	14	4.40	3.61	2017-07-25
08-164	8	278	245	33	0	0.25	0.02	6	0	0	20	0.07	6	382	7.80	3.87	2017-07-29
08-166	9	48	28	20	0	0.08	0.05	26	1	1	7	0.01	6	139	5.00	2.39	2017-07-28
08-168	9	70	38	32	0	0.08	0.04	18	1	1	7	0.01	10	54	5.49	1.74	2017-09-06
08-175	10	324	285	39	0	0.36	0.20	83	0	0	51	0.30	6	548	2.09	5.51	2017-08-18
08-177	10	69	39	30	0	0.10	0.04	16	0	0	10	0.12	7	773	1.38	0.96	2017-09-04
08-179	9	729	694	35	0	0.04	0.57	1004	0	0	0	0.08	17	342	2.59	0.62	2017-08-01

	Dependent variables					Independent variables						Control variables					
08-180	7	53	50	4	0	0.26	0.34	46	0	0	14	0.06	1	752	5.29	1.91	2017-08-03
08-182	11	1333	168	1164	0	0.33	0.12	167	0	1	14	0.02	2	171	3.09	3.95	2017-08-24
08-184	10	107	54	54	0	0.14	0.24	307	0	0	5	0.11	10	1505	4.34	3.79	2017-08-05
08-186	14	600	577	23	0	0.52	0.14	73	0	0	16	0.04	6	98	2.08	2.08	2017-08-20
08-187	10	100	75	25	11	0.12	0.02	19	0	0	12	0.12	1	2847	3.14	4.05	2017-07-24
08-190	12	812	59	753	0	0.30	0.07	12	0	0	12	0.22	8	NA	3.24	2.48	2017-08-21
08-192	8	72	59	13	0	0.30	0.07	10	0	0	8	0.16	13	10	3.94	5.05	2017-08-08
08-193	7	192	186	6	0	0.36	0.06	10	0	0	45	0.13	8	1065	2.53	2.05	2017-08-12
08-202	8	119	94	25	0	0.04	0.03	8	0	0	12	0.03	2	116	1.74	1.25	2017-07-23
08-205	3	8	8	0	0	0.00	0.00	1	0	0	0	0.15	31	667	2.88	4.38	2017-07-28
08-206	7	25	25	0	0	0.00	0.02	1	0	0	0	0.05	8	869	2.50	2.00	2017-07-31

	Dependent variables					Independent variables						Control variables					
08-208	14	249	215	33	5	0.42	0.07	7	0	0	29	0.01	14	48	2.29	2.42	2017-08-13
08-210	11	1134	1118	16	0	0.71	0.04	10	0	0	52	0.09	9	1	2.36	1.36	2017-08-14
08-211	6	109	103	5	0	0.00	0.00	1	0	0	0	0.41	3	6750	2.39	2.91	2017-08-08
08-212	8	90	35	55	0	0.00	0.01	3	0	0	7	0.06	12	807	3.46	3.86	2017-08-07
08-214	11	75	28	46	0	0.33	0.06	10	0	0	9	0.15	3	103	5.33	1.94	2017-09-07
08-218	7	76	32	44	0	0.00	0.01	0	0	0	3	0.06	2	392	2.30	2.01	2017-08-04
08-219	5	20	6	15	0	0.01	0.00	4	0	0	9	0.04	12	NA	1.55	1.43	2017-09-04
09-286	7	59	59	0	0	0.59	0.02	2	0	1	4	0.01	1	159	0.97	0.80	2018-07-20
09-292	4	5	2	3	0	0.00	0.01	0	0	0	0	0.30	12	19919	0.64	6.46	2018-07-27
09-297	4	3	3	0	0	0.00	0.00	0	0	0	0	0.02	2	4009	0.60	1.51	2018-08-02
09-369	1	24	0	24	24	0.00	0.11	4	0	0	0	0.43	1	525	2.37	1.82	2018-07-10

	Dependent variables					Independent variables						Control variables					
09-370	1	0	0	0	0	0.00	0.00	0	0	0	0	0.03	5	350	3.02	0.82	2018-07-12
09-375	3	27	0	27	27	0.12	0.03	4	0	0	16	0.04	17	1899	1.67	1.66	2018-07-19
09-385	4	245	241	5	0	0.40	0.02	2	0	0	10	0.02	1	179	2.79	0.35	2018-07-30
09-392	2	0	0	0	0	0.11	0.01	5	0	0	10	0.12	3	1500	1.95	2.22	2018-08-09
09-393	5	0	0	0	0	0.01	0.07	7	0	0	7	0.05	12	636	1.59	1.23	2018-08-10
09-397	7	1924	112	1811	0	0.37	0.08	44	0	1	18	0.05	2	4603	1.77	1.77	2018-08-14
09-409	15	5116	1000	4116	0	0.56	0.02	0	0	0	15	0.14	3	13195	2.81	4.06	2018-07-03
09-411	3	41	41	0	0	0.37	0.03	3	0	0	7	0.02	4	869	2.41	4.10	2018-07-02
09-427	4	16	7	9	5	0.00	0.03	1	0	0	0	0.11	16	45500	3.46	2.65	2018-07-26
09-428	2	0	0	0	0	0.00	0.00	0	0	0	0	0.03	42	34586	3.09	3.06	2018-07-27
09-430	3	0	0	0	0	0.00	0.02	0	0	0	0	0.03	7	8849	3.58	1.86	2018-07-28

	Dependent variables					Independent variables						Control variables					
09-449	4	2	0	2	0	0.61	0.02	1	0	0	18	0.01	2	1365	0.54	0.13	2018-08-24
09-450	8	0	0	0	0	0.27	0.02	1	0	0	6	0.08	13	8121	1.86	1.64	2018-08-19
09-596	3	9	3	6	0	0.12	0.05	8	0	0	9	0.36	11	9365	1.72	2.58	2019-07-07
09-602	2	2	0	2	0	0.00	0.02	5	0	0	0	0.10	6	2682	2.74	1.56	2019-07-16
09-603	2	4	0	4	0	0.00	0.01	0	0	0	0	0.09	2	12776	1.32	1.80	2019-07-15
09-604	3	24	24	0	0	0.39	0.03	4	0	0	12	0.20	4	2293	2.27	2.18	2019-07-20
09-607	3	29	29	0	0	0.60	0.02	1	0	0	6	0.06	1	350	1.47	2.45	2019-07-21
10-283	11	161	150	11	0	0.00	0.00	0	0	0	0	0.36	4	45500	3.44	1.82	2018-07-15
10-284	14	2544	751	1793	0	0.20	0.02	0	0	0	6	0.02	4	6447	2.29	1.92	2018-07-16
10-301	3	157	0	157	0	0.00	0.01	0	0	0	1	0.29	3	341	0.11	0.25	2018-08-08
10-346	2	0	0	0	0	0.00	0.01	1	0	0	1	0.22	3	300	6.03	0.30	2018-08-11

	Dependent variables					Independent variables						Control variables					
10-349	6	24	0	24	24	0.82	0.03	2	0	0	28	0.00	1	908	1.04	0.37	2018-08-16
10-352	2	47	47	0	0	0.53	0.02	2	0	0	21	0.17	1	202	1.89	0.19	2018-08-19
10-355	6	54	51	3	0	0.53	0.03	1	0	0	8	0.03	10	4772	4.99	0.19	2018-08-23
10-356	11	419	348	71	49	0.43	0.02	0	0	0	10	0.05	2	5334	5.24	0.04	2018-08-24
10-357	3	3	3	0	0	0.37	0.01	1	0	0	7	0.23	2	5107	1.14	0.11	2018-08-25
10-358	14	268	77	191	0	0.01	0.01	1	0	0	0	0.01	4	45500	3.10	0.61	2018-08-28
10-359	5	31	31	0	0	0.00	0.00	0	0	0	0	0.11	2	45500	5.06	0.87	2018-08-29
10-387	3	346	0	346	0	0.55	0.04	14	1	1	23	0.00	3	21	1.62	0.69	2018-08-24
10-388	9	664	51	613	0	0.01	0.23	70	0	0	9	0.02	2	39099	1.71	3.15	2018-08-04
10-389	5	103	102	1	0	0.75	0.03	1	0	0	16	0.01	2	1932	0.53	3.61	2018-08-05
10-398	6	199	192	7	0	0.52	0.03	2	0	0	16	0.04	1	404	2.37	1.67	2018-08-17

	Dependent variables					Independent variables						Control variables					
10-399	10	888	314	575	0	0.75	0.05	11	1	1	11	0.04	4	4561	1.39	1.62	2018-08-18
10-400	4	0	0	0	0	0.02	0.18	82	0	0	1	0.17	2	10663	3.29	0.59	2018-08-26
10-402	6	1321	37	1284	0	0.06	0.04	4	0	0	5	0.18	1	26910	1.62	0.28	2018-08-21
10-406	5	75	75	0	0	0.52	0.02	2	0	0	26	0.09	1	10979	1.83	0.56	2018-08-25
10-412	4	174	174	0	0	0.53	0.03	3	1	1	7	0.01	2	526	1.43	3.78	2018-07-06
10-416	2	0	0	0	0	0.54	0.02	1	0	0	4	0.00	1	415	0.20	2.67	2018-07-10
10-433	8	53	46	6	0	0.88	0.02	0	0	0	4	0.00	1	273	1.63	0.37	2018-08-02
10-435	6	187	187	0	0	0.30	0.02	1	1	1	12	0.01	15	4784	2.15	1.74	2018-08-05
10-438	5	96	96	0	0	0.75	0.03	0	0	0	2	0.02	2	45500	1.13	1.44	2018-08-04
10-444	5	24	16	8	4	0.47	0.02	1	0	0	7	0.01	6	1226	2.90	1.42	2018-08-16
10-445	9	16	16	0	0	0.67	0.01	0	0	0	6	0.02	4	5511	2.33	1.39	2018-08-14

	Dependent variables					Independent variables						Control variables					
10-452	7	62	6	56	53	0.05	0.03	1	0	0	8	0.08	1	905	0.29	0.10	2018-08-26
10-453	5	90	85	4	0	0.57	0.02	1	0	0	4	0.01	2	592	4.33	1.27	2018-08-13
10-597	6	94	66	29	3	0.01	0.26	104	0	0	2	0.07	3	835	2.96	3.68	2019-07-08
10-598	4	34	26	8	0	0.00	0.02	0	0	0	0	0.11	5	14875	1.47	2.43	2019-07-09
11-342	0	0	0	0	0	0.00	0.00	0	0	0	0	0.06	18	1663	49.74	0.73	2018-08-06
11-470	2	1	0	1	0	0.00	0.03	1	0	0	6	0.10	19	760	7.77	3.28	2019-07-14
11-478	1	0	0	0	0	0.00	0.06	5	0	0	0	0.03	5	101	9.87	2.24	2019-07-24
11-480	0	0	0	0	0	0.00	0.00	0	0	0	0	0.13	8	2467	8.82	1.59	2019-07-27
11-481	0	0	0	0	0	0.00	0.01	0	0	0	0	0.06	13	1448	6.75	1.69	2019-07-28
11-482	0	0	0	0	0	0.00	0.00	0	0	0	0	0.15	7	1815	12.55	1.57	2019-07-29
11-483	1	6	0	6	0	0.00	0.00	0	0	0	0	0.14	9	311	7.06	1.75	2019-07-30

	Dependent variables					Independent variables						Control variables					
11-484	4	48	0	48	0	0.00	0.03	5	0	0	16	0.07	12	1624	6.62	2.74	2019-08-01
11-486	0	0	0	0	0	0.03	0.01	1	0	0	9	0.00	8	163	7.38	1.44	2019-08-03
11-487	2	0	0	0	0	0.03	0.01	1	0	0	9	0.01	3	281	7.02	1.44	2019-08-04
11-488	0	0	0	0	0	0.06	0.02	2	0	0	17	0.11	15	5537	7.78	1.54	2019-08-05
11-489	1	0	0	0	0	0.00	0.01	1	0	0	5	0.06	19	3742	9.89	1.02	2019-08-06
11-491	1	24	0	24	24	0.02	0.03	3	0	0	4	0.04	6	262	7.07	1.75	2019-08-10
11-493	1	203	0	203	0	0.00	0.05	2	0	0	1	0.23	4	87	3.98	1.58	2019-08-12
11-495	1	0	0	0	0	0.00	0.02	4	0	0	3	0.00	24	54	7.24	2.46	2019-08-16
11-499	0	0	0	0	0	0.00	0.01	2	0	0	0	0.11	28	2451	22.88	0.11	2019-08-20
11-503	0	0	0	0	0	0.00	0.00	0	0	0	0	0.03	5	453	9.41	0.67	2019-08-25
11-508	0	0	0	0	0	0.00	0.00	0	0	0	0	0.03	30	229	18.63	4.34	2019-07-04

	Dependent variables					Independent variables						Control variables					
11-509	1	0	0	0	0	0.00	0.00	0	0	0	0	0.04	36	188	12.44	4.86	2019-07-05
11-541	0	0	0	0	0	0.00	0.00	0	0	0	0	0.07	42	891	14.53	3.78	2019-08-20
11-546	1	1279	0	1279	0	0.00	0.00	0	0	0	0	0.01	4	42	7.23	1.32	2019-08-24
11-547	0	0	0	0	0	0.00	0.00	0	0	0	0	0.04	19	1174	11.73	1.04	2019-08-26
11-556	2	0	0	0	0	0.00	0.46	184	0	0	0	0.05	3	264	7.65	4.98	2019-07-07
11-557	2	0	0	0	0	0.00	0.05	1	0	0	0	0.10	17	535	9.48	4.99	2019-07-08
11-589	0	0	0	0	0	0.00	0.00	0	0	0	0	0.00	36	13	12.91	4.98	2019-08-23
11-627	2	0	0	0	0	0.00	0.00	0	0	0	0	0.19	7	477	9.43	3.07	2019-08-18
11-628	1	0	0	0	0	0.00	0.00	0	0	0	0	0.01	7	4	11.71	3.84	2019-08-19
11-629	0	0	0	0	0	0.00	0.00	0	0	0	0	0.09	16	462	9.55	2.15	2019-08-20
11-630	1	0	0	0	0	0.00	0.00	0	0	0	0	0.18	21	4	13.11	0.64	2019-08-23

	Dependent variables					Independent variables						Control variables					
11-633	1	37	0	37	0	0.00	0.06	0	0	0	0	0.06	10	27	10.61	4.49	2019-08-26
12-460	6	374	35	339	0	0.00	0.81	2677	0	0	6	0.12	12	66	4.87	3.39	2019-06-29
12-462	6	636	21	616	0	0.01	0.83	3705	0	0	0	0.04	6	48	7.30	2.89	2019-06-30
12-466	3	3	3	0	0	0.00	0.01	3	1	0	0	0.01	77	256	49.47	2.55	2019-07-06
12-473	1	0	0	0	0	0.00	0.01	1	0	0	0	0.12	16	1914	26.19	2.80	2019-07-13
12-504	3	0	0	0	0	0.00	0.00	0	0	0	0	0.04	50	340	61.45	1.38	2019-08-26
12-528	0	0	0	0	0	0.00	0.00	0	0	0	0	0.03	12	53	52.99	5.93	2019-08-01
12-529	0	0	0	0	0	0.00	0.00	0	0	0	0	0.01	28	48	19.57	2.48	2019-08-02
12-530	0	0	0	0	0	0.00	0.00	0	0	0	0	0.02	15	47	19.59	1.70	2019-08-03
12-531	3	0	0	0	0	0.00	0.00	0	0	0	0	0.02	56	323	46.44	1.84	2019-08-04
12-533	3	0	0	0	0	0.00	0.01	0	0	0	0	0.01	6	NA	20.38	0.21	2019-08-10

	Dependent variables					Independent variables						Control variables					
12-534	1	0	0	0	0	0.00	0.01	0	0	0	0	0.06	19	182	23.06	0.17	2019-08-09
12-535	1	0	0	0	0	0.00	0.01	0	0	0	0	0.01	40	49	20.50	0.10	2019-08-08
12-537	1	786	0	786	0	0.00	0.01	1	0	0	0	0.02	4	63	8.87	1.31	2019-08-13
12-636	4	22	0	22	0	0.00	0.10	148	0	0	0	0.07	10	20	14.08	1.47	2019-07-02
12-637	0	0	0	0	0	0.00	0.16	27	0	0	0	0.06	12	152	13.88	0.74	2019-07-04
12-640	0	0	0	0	0	0.00	0.58	199	0	0	0	0.13	9	716	12.67	0.40	2019-07-07
12-641	0	0	0	0	0	0.00	0.44	233	0	0	0	0.29	5	909	12.02	0.62	2019-07-08
12-642	3	0	0	0	0	0.00	0.47	361	0	0	0	0.24	16	28	12.66	1.33	2019-07-11
12-644	3	3	0	3	0	0.00	0.17	43	0	0	4	0.05	12	114	12.58	1.65	2019-07-13
12-646	3	49	0	49	0	0.07	0.42	108	0	0	48	0.02	16	120	9.09	1.66	2019-07-15
12-648	1	0	0	0	0	0.06	0.38	266	0	0	37	0.17	8	285	8.84	0.97	2019-07-18

	Dependent variables					Independent variables						Control variables					
12-649	2	0	0	0	0	0.00	0.42	95	0	0	0	0.33	16	1806	4.03	1.69	2019-07-19
12-651	1	2	0	2	0	0.03	0.27	108	0	0	12	0.02	5	43	6.92	2.29	2019-07-21
12-653	6	0	0	0	0	0.00	0.00	0	0	0	0	0.03	13	450	10.51	0.10	2019-07-27
12-654	1	0	0	0	0	0.00	0.00	0	0	0	0	0.13	41	1049	46.35	0.17	2019-07-25
12-655	3	66	38	28	28	0.00	0.00	0	0	0	0	0.02	11	27	31.06	0.16	2019-07-26
12-656	2	28	0	28	28	0.00	0.22	69	0	0	0	0.07	12	328	17.93	0.10	2019-07-30
12-657	2	27	0	27	0	0.00	0.50	252	0	0	1	0.05	9	850	15.61	0.17	2019-07-28
12-658	0	0	0	0	0	0.00	0.00	0	0	0	0	0.12	38	444	14.92	6.91	2019-08-05
12-659	3	0	0	0	0	0.00	0.00	0	0	0	0	0.02	53	75	27.50	8.04	2019-08-03
12-662	1	0	0	0	0	0.00	0.00	0	0	0	0	0.13	138	1158	51.52	0.07	2019-08-09
12-665	3	2	2	0	0	0.00	0.00	0	0	0	0	0.05	41	192	62.41	0.32	2019-08-12

	Dependent variables					Independent variables						Control variables					
12-670	3	173	0	173	0	0.00	0.76	1207	0	0	0	0.03	8	87	7.90	0.83	2019-08-15
12-671	2	22	0	22	0	0.00	0.13	147	0	0	1	0.02	20	107	17.30	0.82	2019-08-16
12-672	4	25	0	25	0	0.00	0.71	1342	0	0	0	0.19	14	735	10.61	0.84	2019-08-17
12-675	1	0	0	0	0	0.00	0.01	1	0	0	0	0.07	51	425	36.47	0.24	2019-08-20
12-676	0	0	0	0	0	0.00	0.12	3	0	0	0	0.18	12	362	6.43	2.56	2019-08-23
12-677	1	0	0	0	0	0.00	0.00	0	0	0	0	0.18	65	2945	27.87	2.99	2019-08-25
13-514	1	0	0	0	0	0.00	0.05	0	0	0	0	0.18	19	26	11.65	0.82	2019-07-12
13-516	1	0	0	0	0	0.00	0.02	0	0	0	0	0.04	41	456	43.35	0.46	2019-07-14
13-517	0	0	0	0	0	0.00	0.10	38	0	1	0	0.01	28	141	10.38	0.67	2019-07-16
13-519	4	1	0	1	0	0.00	0.01	0	0	0	0	0.08	36	1411	9.13	0.70	2019-07-15
13-521	0	0	0	0	0	0.00	0.00	0	0	0	0	0.00	9	21	26.87	1.15	2019-07-20

	Dependent variables					Independent variables						Control variables					
13-523	1	0	0	0	0	0.00	0.00	0	0	0	0	0.03	127	2155	34.96	1.09	2019-07-23
13-525	3	0	0	0	0	0.00	0.00	0	0	0	0	0.07	3	5083	24.75	2.73	2019-07-26
13-526	2	0	0	0	0	0.00	0.00	0	0	0	0	0.05	28	7555	18.34	5.21	2019-07-25
13-560	2	0	0	0	0	0.00	0.08	50	0	0	0	0.29	33	2087	10.00	0.61	2019-07-13
13-562	1	0	0	0	0	0.00	0.09	1	0	0	0	0.10	7	1160	3.86	0.74	2019-07-15
13-563	1	0	0	0	0	0.00	0.04	0	0	0	0	0.20	3	541	2.99	2.73	2019-07-16
13-565	1	0	0	0	0	0.00	0.01	0	0	0	0	0.01	21	2478	16.87	1.97	2019-07-18
13-566	2	0	0	0	0	0.00	0.05	0	0	0	0	0.10	21	2893	23.62	0.96	2019-07-20
13-568	0	0	0	0	0	0.00	0.00	0	0	0	0	0.15	38	10820	20.40	0.12	2019-07-23
13-569	1	0	0	0	0	0.00	0.00	0	0	0	0	0.05	34	6048	18.64	0.35	2019-07-24
13-587	0	0	0	0	0	0.00	0.03	16	0	0	0	0.15	6	72	4.70	1.61	2019-08-17

	Dependent variables					Independent variables						Control variables					
14-585	3	835	0	835	0	0.00	0.02	0	0	0	0	0.14	1	680	2.20	1.92	2019-08-12
17-038	3	7	7	0	0	0.00	0.02	0	0	0	0	0.05	9	128	8.82	1.25	2017-07-14
17-043	4	19	19	0	0	0.00	0.00	0	0	0	0	0.17	13	498	9.13	0.97	2017-07-20
17-050	3	21	0	21	0	0.00	0.26	98	0	0	0	0.04	1	21	2.26	2.92	2017-08-11
17-054	1	0	0	0	0	0.00	0.04	0	0	0	0	0.14	5	1105	6.68	0.70	2017-07-31
17-056	3	35	14	21	0	0.00	0.00	1	0	0	0	0.21	6	1494	5.51	0.26	2017-08-03
17-059	5	21	11	10	0	0.13	0.01	5	0	0	13	0.04	3	137	5.73	2.76	2017-08-07
17-069	4	2	0	2	0	0.00	0.01	3	0	0	1	0.04	47	325	9.72	2.40	2017-07-12
17-070	4	4	2	1	0	0.01	0.01	4	1	1	1	0.03	50	493	8.60	0.72	2017-07-29
17-072	3	11	11	0	0	0.06	0.01	4	0	0	15	0.09	3	338	8.80	0.25	2017-08-05
17-075	3	1	0	1	0	0.04	0.03	19	0	0	12	0.02	9	53	4.43	2.53	2017-08-07

	Dependent variables					Independent variables						Control variables					
17-076	1	0	0	0	0	0.03	0.00	2	0	0	5	0.05	5	210	4.56	0.54	2017-07-30
17-079	2	2	0	2	0	0.00	0.00	0	0	0	0	0.02	10	131	5.30	1.14	2017-07-24
17-081	3	4	0	4	4	0.00	0.00	0	0	0	0	0.04	26	276	8.99	2.36	2017-07-25
17-084	3	5	5	0	0	0.00	0.00	0	0	0	0	0.07	25	306	8.39	3.39	2017-07-21
17-087	5	27	27	0	0	0.00	0.04	16	0	0	8	0.10	3	146	3.66	3.67	2017-07-11
17-088	7	33	11	23	0	0.01	0.06	59	0	0	8	0.06	30	241	5.84	4.51	2017-07-22
17-090	3	1	1	0	0	0.00	0.02	9	0	0	13	0.01	6	16	5.16	3.49	2017-08-22
17-092	1	5	5	0	0	0.00	0.01	8	0	1	1	0.04	58	294	9.38	2.79	2017-07-18
17-093	2	19	19	0	0	0.01	0.02	9	0	0	4	0.05	5	117	10.33	2.86	2017-07-19
17-094	3	259	257	2	2	0.01	0.03	11	0	0	23	0.04	2	65	6.10	2.87	2017-08-26
17-098	20	304	68	236	0	0.01	0.10	56	1	1	12	0.00	2	6	8.39	5.16	2017-08-24

	Dependent variables					Independent variables						Control variables					
17-105	9	224	216	9	6	0.08	0.03	55	1	0	14	0.04	2	87	4.16	2.01	2017-08-27
17-107	5	592	584	8	0	0.03	0.38	19	0	0	9	0.07	1	136	5.14	0.16	2017-08-30
17-109	12	183	76	107	0	0.02	0.06	33	1	1	2	0.01	19	27	7.16	3.59	2017-07-10
17-110	5	50	49	1	0	0.03	0.05	20	0	0	53	0.14	8	227	4.40	4.63	2017-07-13
17-111	4	37	37	0	0	0.00	0.03	26	0	0	1	0.31	19	1487	6.15	3.80	2017-07-10
17-114	8	33	29	4	4	0.00	0.02	18	0	0	1	0.08	25	620	9.23	3.68	2017-07-10
17-117	6	57	51	6	0	0.02	0.05	21	1	0	1	0.05	85	960	8.23	4.82	2017-07-14
17-119	10	126	31	95	13	0.01	0.06	29	0	0	5	0.08	7	233	6.54	4.24	2017-07-10
17-123	3	34	29	5	0	0.01	0.01	26	0	0	8	0.02	7	53	15.45	4.42	2017-07-11
18-324	15	5115	285	4830	75	0.03	0.07	40	1	0	7	0.04	50	4193	19.74	0.39	2018-07-13
18-335	2	0	0	0	0	0.00	0.00	0	0	0	5	0.01	53	698	14.31	0.39	2018-07-23

	Dependent variables					Independent variables						Control variables					
18-336	2	0	0	0	0	0.00	0.00	0	0	0	0	0.45	11	2689	7.77	0.07	2018-07-28
18-337	3	370	0	370	0	0.02	0.06	50	1	1	4	0.00	54	306	20.93	0.03	2018-08-02
18-338	3	0	0	0	0	0.03	0.01	2	0	0	3	0.05	20	1730	27.63	0.01	2018-07-30
18-339	10	20	2	18	0	0.02	0.06	50	1	1	4	0.00	24	14	20.95	0.00	2018-07-31
18-505	1	1	0	1	0	0.00	0.00	0	0	0	6	0.02	6	301	14.97	2.07	2019-06-30
18-552	2	7	0	7	0	0.00	0.11	3	0	0	0	0.21	10	1221	8.83	2.55	2019-07-02
18-553	3	0	0	0	0	0.00	0.00	0	0	0	0	0.03	3	143	11.01	2.44	2019-07-04
18-593	0	0	0	0	0	0.00	0.00	0	0	0	0	0.08	17	3454	10.60	2.25	2019-07-02

Tableau 21. Results for the negative binomial and ordinal logistic regression models.

Variable	β	Standard error	*z*-value	*p*-value *	IRR or OR (95%CI) *
Number of detections (n = 283)					
(Intercept)	-0.74	0.70	-1.06	0.3	0.48 (0.12-1.85)
Agricultural fraction	0.01	0.003	3.86	**< 0.001**	**1.11** (1.05-.180)
Urban fraction	0.02	0.003	5.06	**< 0.001**	**1.17** (1.10-1.25)
WWTPs presence	1.00	0.17	5.84	**< 0.001**	**2.72** (1.95-3.85)
Area ratio	-0.92	0.55	-1.68	0.1	0.40 (0.13-1.19)
Lake depth (ln)	-0.003	0.06	-0.05	1.0	1.00 (0.89-1.12)
Residence time (ln)	0.03	0.03	1.10	0.3	1.03 (0.98-1.08)
Mean slope (ln)	-0.14	0.06	-2.18	**0.03**	**0.87** (0.77-0.99)
Precipitation	0.10	0.03	3.33	**< 0.001**	**1.10** (1.04-1.17)
Sampling date	0.008	0.003	2.78	**0.005**	**1.01** (1.00-1.01)
Total TrOCs sum (n = 284)					
Agricultural fraction	0.02	0.01	3.49	**< 0.001**	**1.26** (1.11-1.44)
Urban fraction	0.05	0.01	5.99	**< 0.001**	**1.58** (1.36-1.84)
WWTPs presence	1.94	0.41	4.74	**< 0.001**	**6.95** (3.12-15.5)
Area ratio	-1.14	1.24	-0.92	0.4	0.32 (0.03-3.64)
Lake depth (ln)	-0.31	0.13	-2.43	**0.02**	**0.73** (0.57-0.94)
Residence time (ln)	0.01	0.06	0.13	0.9	1.01 (0.89-1.14)
Mean slope (ln)	-0.28	0.15	-1.91	0.1	0.76 (0.57-1.01)
Precipitation	0.24	0.07	3.49	**< 0.001**	**1.27** (1.11-1.45)
Sampling date	0.01	0.01	1.65	0.1	1.01 (1.00-1.02)
Pesticides' sum (n = 284)					
Agricultural fraction	0.05	0.01	6.04	**< 0.001**	**1.58** (1.36-1.83)
Urban fraction	0.02	0.01	2.26	**0.02**	**1.19** (1.02-1.39)
Area ratio	-1.54	1.37	-1.12	0.3	0.21 (0.01-3.15)
Lake depth (ln)	-0.21	0.14	-1.51	0.1	0.81 (0.62-1.06)
Residence time (ln)	0.02	0.07	0.36	0.7	1.02 (0.90-1.17)
Mean slope (ln)	-0.07	0.16	-0.42	0.7	0.94 (0.69-1.27)
Precipitation	0.29	0.07	4.01	**< 0.001**	**1.34** (1.16-1.55)
Sampling date	0.01	0.01	1.22	0.2	1.01 (0.99-1.02)

Variable	β	Standard error	*z*-value	*p*-value *	IRR or OR (95%CI) *
PPCPs and additives' sum (n = 284)					
Urban fraction	0.05	0.01	6.13	**< 0.001**	**1.57** (1.36-1.82)
WWTPs presence	1.72	0.54	3.17	**0.002**	**5.57** (1.92-16.1)
Hospitals presence	0.14	0.64	0.22	0.8	1.15 (0.33-4.00)
Area ratio	-1.08	1.19	-0.91	0.4	0.34 (0.03-3.51)
Lake depth (ln)	-0.20	0.13	-1.53	0.1	0.82 (0.63-1.06)
Residence time (ln)	-0.01	0.06	-0.18	0.9	0.99 (0.88-1.12)
Mean slope (ln)	-0.33	0.14	-2.34	**0.02**	**0.72** (0.54-0.95)
Precipitation	0.17	0.07	2.48	**0.01**	**1.19** (1.04-1.36)
Sampling date	0.01	0.01	1.34	0.2	1.01 (0.99-1.02)
Antibiotics' sum (n = 284)					
Livestock density	0.02	0.02	0.84	0.4	1.01 (0.98-1.05)
WWTPs presence	0.66	0.63	1.06	0.3	1.94 (0.57-6.65)
Population density	-3.00E-04	8.00E-04	-0.41	0.7	1.00 (1.00-1.00)
Area ratio	-1.02	1.89	-0.54	0.6	0.36 (0.01-14.7)
Lake depth (ln)	-0.10	0.21	-0.48	0.6	0.90 (0.60-1.36)
Residence time (ln)	-0.01	0.10	-0.07	0.9	0.99 (0.82-1.20)
Mean slope (ln)	-0.26	0.22	-1.16	0.2	0.77 (0.50-1.19)
Precipitation	-0.01	0.12	-0.09	0.9	0.99 (0.78-1.25)
Sampling date	-0.01	0.01	-1.12	0.3	0.99 (0.97-1.01)

* Values shown in bold represent a statistically significant association.

Tableau 22. Information on Lake Pulse lakes not included in the study of TrOCs.

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
04-573	Dalts'an Jìthìnùu Vàn, NT	67.34958	-134.923101	0.8	low	0.00	0.01	0	0	0	0
04-574	NA, NT	67.417758	-133.922557	0.1	low	0.00	0.02	0	0	0	0
04-575	NA, NT	67.381817	-134.040849	0.2	moderate	0.00	0.05	0	0	0	0
04-576	Chii Eche i i Van, NT	67.468001	-133.763637	0.4	moderate	0.00	0.01	0	0	0	0
04-578	Boot Lake, NT	68.350907	-133.708655	0.2	moderate	0.00	0.05	35	0	1	0
04-581	NA, NT	67.432054	-134.851562	0.4	moderate	0.00	0.06	32	0	0	0
04-582	NA, NT	68.335154	-133.680514	0.3	moderate	0.00	0.02	0	0	0	0
04-611	NA, NT	60.063822	-116.847789	0.4	low	0.00	0.00	0	0	0	0
04-620	NA, NT	61.998774	-116.330501	0.7	low	0.00	0.01	0	0	0	0
04-621	NA, NT	61.930612	-116.520168	0.4	moderate	0.00	0.04	0	0	0	0
04-622	NA, BC	59.798928	-122.523299	0.1	moderate	0.00	0.03	0	0	0	0
06-036	Lac des Rapides, QC	50.313788	-66.428761	5.8	low	0.00	0.00	0	0	0	0
06-066	NA, QC	49.869455	-68.732763	0.4	low	0.00	0.01	0	0	0	0
06-071	Lac Parenthèses, QC	50.365341	-68.818863	1.9	moderate	0.00	0.05	0	0	0	0
06-080	Lac Jérôme, QC	48.248292	-69.637829	0.2	low	0.00	0.01	2	0	0	0
06-095	Lac Simoncouche, QC	48.231271	-71.251055	0.8	low	0.00	0.02	0	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
06-096	Lac Saint-Charles, QC	46.948182	-71.392515	3.6	moderate	0.00	0.10	73	1	0	2
06-100	Lac Élie-Gagnon, QC	48.721602	-71.671653	0.1	high	0.21	0.02	29	0	0	2
06-101	Lac Maupertuis, QC	50.418743	-71.734219	14.2	low	0.00	0.00	0	0	0	0
06-124	Lac de l'Achigan, QC	45.943917	-73.977427	5.5	moderate	0.00	0.06	31	0	0	0
06-125	Lac Croche, QC	45.992729	-74.00935	0.2	low	0.00	0.00	7	0	0	0
06-131	Lac Rond, QC	45.948099	-74.143866	0.2	high	0.00	0.60	575	0	0	0
06-140	Lac Daine, QC	49.87207	-75.664784	2.3	low	0.00	0.00	0	0	0	0
06-141	Lac Pelletier, QC	48.214044	-79.052766	3.0	high	0.00	0.29	263	1	0	0
06-142	Lac Manitou, QC	45.875279	-75.956424	0.6	moderate	0.00	0.00	1	0	0	3
06-146	Lac Blue Sea, QC	46.192548	-76.057327	14.6	moderate	0.00	0.04	11	0	0	1
06-156	Wabun Lake, ON	45.226736	-76.831453	0.5	low	0.00	0.00	0	0	0	0
06-159	Mink Lake, ON	45.564777	-77.04943	5.7	moderate	0.02	0.05	12	0	0	10
06-161	Fraser Lake, ON	45.190967	-77.651071	2.3	moderate	0.00	0.05	6	0	0	3
06-165	Lac Obalski, QC	48.753092	-77.960512	18.1	moderate	0.01	0.02	13	1	1	1
06-171	Lac Macamic, QC	48.798152	-78.975731	45.9	moderate	0.01	0.01	3	1	0	2
06-178	Gull Lake, ON	44.920063	-79.359743	1.4	moderate	0.00	0.17	114	0	0	0
06-198	Dora Lake, ON	49.186723	-80.970066	0.6	high	0.24	0.02	2	0	0	2
06-199	Lillabelle Lake, ON	49.109138	-81.032121	1.8	high	0.24	0.19	133	1	1	2

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
06-253	Burnt Berry Pond, NL	49.451178	-56.188657	1.6	low	0.00	0.00	0	0	0	0
06-255	Gull Pond, NL	47.46447	-52.976425	0.5	moderate	0.04	0.15	0	0	0	9
06-256	Three Island Pond, NL	47.510293	-52.901442	0.9	high	0.00	0.29	35	0	0	0
06-257	Octagon Pond, NL	47.524518	-52.881807	0.5	high	0.00	0.67	541	0	0	0
06-259	Millers Pond, NL	47.623691	-52.825964	0.2	high	0.00	0.41	96	0	0	12
06-260	Great Pond, NL	47.664336	-52.76537	0.3	moderate	0.00	0.13	58	0	0	13
06-262	Stern Pond, NL	47.129687	-55.514258	0.3	moderate	0.00	0.13	3	0	0	0
06-263	Freshwater Pond, NL	47.102985	-55.260614	9.7	low	0.00	0.00	0	0	0	0
06-264	Sandy Cove Pond, NL	48.64754	-53.729453	0.3	moderate	0.00	0.15	35	0	0	0
06-265	Cobbs Pond, NL	48.971275	-54.629757	0.3	high	0.00	0.34	161	0	0	0
06-266	River Pond, NL	47.915888	-55.586692	1.5	low	0.00	0.00	0	0	0	0
06-267	Tippings Pond, NL	48.930281	-57.880527	0.2	low	0.00	0.05	59	0	0	0
06-268	Grand Pond, NL	51.087718	-56.860944	2.5	low	0.00	0.01	1	0	0	0
06-270	Curls Pond, NL	52.413575	-56.01578	2.8	low	0.00	0.01	0	0	0	0
06-271	Denmark Lake, ON	49.138532	-91.218791	0.2	low	0.00	0.00	0	0	0	0
06-272	NA, ON	49.453078	-92.08314	0.7	moderate	0.00	0.04	0	0	0	0
06-273	Round Lake, ON	49.778297	-94.444095	0.2	high	0.00	0.37	158	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
06-275	Whitemouth Lake, MB	49.230934	-95.682275	69.8	low	0.01	0.00	0	0	0	0
06-289	Barbe Lake, MB	54.282254	-101.474724	1.9	low	0.00	0.00	0	0	0	0
06-294	NA, MB	55.892985	-98.996064	0.7	low	0.00	0.03	0	0	0	0
06-295	Eden Lake, MB	56.624606	-100.17423	67.1	low	0.00	0.00	0	0	0	0
06-303	Gull Lake, MB	50.409666	-96.514633	1.2	moderate	0.00	0.19	28	0	0	0
06-305	L302, ON	49.674081	-93.762805	0.2	low	0.00	0.01	0	0	0	0
06-306	L223, ON	49.698298	-93.70868	0.3	low	0.00	0.00	0	0	0	0
06-307	L227, ON	49.687897	-93.688896	0.1	low	0.00	0.00	0	0	0	0
06-308	L373, ON	49.74382	-93.799896	0.3	low	0.00	0.03	0	0	0	0
06-309	Rob Lake, ON	50.401284	-91.254248	0.8	high	0.00	0.02	0	0	0	0
06-310	Ponsford Lake, ON	51.506057	-90.339539	0.7	high	0.00	0.09	4	0	0	0
06-311	L224, ON	49.690256	-93.717327	0.3	low	0.00	0.00	0	0	0	0
06-312	Botsford Lake, ON	50.147688	-91.647757	14.6	low	0.00	0.00	0	0	0	0
06-313	Pakashkan Lake, ON	49.337978	-90.271499	50.5	low	0.00	0.00	0	0	0	0
06-314	O'Sullivan Lake, ON	50.407677	-87.080146	41.6	low	0.00	0.00	0	1	0	0
06-455	NA, NL	47.850967	-55.765996	0.0	low	0.00	0.00	0	0	0	0
06-456	Kississing lake, MB	55.131569	-101.446955	266.3	low	0.00	0.01	0	0	0	0
07-001	Whitlocks Pond, PE	46.349582	-62.52702	0.7	moderate	0.18	0.02	4	0	0	6
07-004	Mosleys Pond, NS	45.92193	-64.087807	0.3	low	0.00	0.00	1	0	0	0
07-011	Wiles Lake, NS	44.373332	-64.574322	1.0	moderate	0.00	0.16	42	0	0	2

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
07-014	Tupper Lake, NS	45.017746	-64.589997	0.3	moderate	0.00	0.13	7	0	0	0
07-016	Folly Lake, NB	46.036878	-64.64533	0.2	moderate	0.00	0.25	29	0	0	0
07-019	Caribou Lake, NS	44.525535	-64.54906	2.8	low	0.00	0.03	5	0	0	0
07-021	Kent Lake, NB	46.636229	-65.117947	0.2	low	0.00	0.01	1	0	0	0
07-025	French Lake, NS	43.639494	-65.707216	5.6	low	0.00	0.01	1	0	0	0
07-028	Everitts Lake, NS	44.448946	-65.861249	0.5	moderate	0.00	0.07	3	0	0	1
07-032	Clare Lake, NS	44.193053	-66.103988	0.5	moderate	0.00	0.08	21	0	0	3
07-046	Oromocto Lake, NB	45.586373	-67.003961	41.6	low	0.00	0.02	3	0	0	1
07-047	Magaguadavic Lake, NB	45.716645	-67.20662	27.0	low	0.00	0.01	0	0	0	0
07-049	NA, NB	46.897616	-67.432087	0.2	moderate	0.04	0.04	8	0	0	2
07-051	Blue Bell Lake, NB	46.950127	-67.537914	0.1	low	0.00	0.03	15	0	0	0
07-055	Lake Edward, NB	46.921096	-67.65543	0.2	high	0.28	0.04	6	0	0	1
07-057	Piries Lake, NB	46.936727	-67.758306	0.2	high	0.26	0.02	4	0	0	0
07-222	Big Lake, NS	45.710947	-63.710239	1.3	moderate	0.01	0.03	4	0	0	4
07-223	Kennedy Lake, NS	45.716514	-63.228122	0.1	moderate	0.02	0.04	7	0	0	4
07-224	Folly Lake, NS	45.537496	-63.544962	0.9	low	0.00	0.04	3	0	0	0
07-225	Little Dyke Lake, NS	45.385397	-63.560296	0.1	high	0.03	0.65	67	0	0	3
07-226	Trout Lake, NS	45.572019	-63.488455	0.2	low	0.00	0.01	0	0	0	0
07-227	Shortts Lake, NS	45.219083	-63.31812	2.1	moderate	0.01	0.10	32	0	0	4

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
07-232	Pigott Lake, NS	44.929835	-63.876439	0.9	moderate	0.00	0.15	17	0	0	1
07-233	Springfield Lake, NS	44.808913	-63.742615	0.8	high	0.00	0.44	336	0	0	0
07-234	Halfway Lake, NS	44.745336	-63.785165	0.1	high	0.00	0.68	395	0	0	0
07-235	Kearney Lake, NS	44.696009	-63.698055	0.6	moderate	0.00	0.24	455	0	0	0
07-238	First Chain Lake, NS	44.637112	-63.651611	0.3	high	0.00	0.53	65	0	0	0
07-239	Lake Banook, NS	44.692693	-63.553527	1.5	high	0.00	0.87	1028	0	0	0
07-240	Morris Lake, NS	44.651541	-63.497262	1.7	high	0.00	0.65	1115	0	0	0
07-241	Bissett Lake, NS	44.655567	-63.469954	0.9	high	0.00	0.89	2091	0	0	0
07-242	Loon Lake, NS	44.702019	-63.504617	0.8	high	0.00	0.62	312	0	0	0
07-244	West Pogue Lake, NS	45.228384	-62.762945	0.2	low	0.00	0.00	1	0	0	0
07-245	Pringle Lake, NS	45.376743	-61.94841	0.6	moderate	0.00	0.04	7	0	0	5
07-246	Gaspereaux Lake, NS	45.556219	-62.056197	0.9	moderate	0.00	0.04	25	0	0	7
07-249	Embrees Pond, NS	45.622179	-61.362123	0.1	moderate	0.00	0.17	146	0	0	0
07-252	Johnson Lake, NS	46.171199	-60.318258	0.4	low	0.00	0.02	9	0	0	0
08-120	Lac des Chicots, QC	46.796216	-72.523542	0.7	moderate	0.25	0.09	88	0	0	6
08-138	Lac Écho, QC	45.891675	-74.028795	1.6	high	0.00	0.26	178	0	0	0
08-143	Constance Lake, ON	45.409983	-75.979067	1.5	moderate	0.06	0.09	40	0	0	9
08-144	Charleston Lake, ON	44.544584	-76.047291	27.5	low	0.09	0.04	15	0	0	8

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
08-148	Upper Rock Lake, ON	44.494194	-76.413815	0.8	low	0.00	0.03	8	0	0	1
08-149	Newboro Lake, ON	44.630897	-76.318336	16.8	low	0.05	0.04	7	0	0	4
08-150	Round Lake, ON	44.414982	-76.429486	0.6	low	0.01	0.06	24	0	0	6
08-152	Christie Lake, ON	44.603507	-76.467922	0.3	low	0.00	0.01	1	0	0	0
08-153	Lac Green, QC	45.651003	-76.495042	0.6	low	0.17	0.01	7	0	0	8
08-157	Camden Lake, ON	44.409097	-76.857259	6.2	moderate	0.07	0.03	12	0	0	14
08-162	Little Lake, ON	44.046054	-77.824936	0.7	high	0.34	0.15	60	0	0	7
08-163	Seymour Lake, ON	44.380086	-77.814912	7.5	moderate	0.09	0.05	26	1	1	8
08-169	Lake Scugog, ON	44.191502	-78.809868	69.4	moderate	0.28	0.08	48	1	1	18
08-172	NA, ON	42.992977	-80.607949	0.1	high	0.60	0.04	24	0	0	78
08-183	NA, ON	43.747219	-79.735133	0.3	high	0.00	0.96	3539	0	0	0
08-185	Orr Lake, ON	44.608247	-79.802979	3.2	moderate	0.04	0.15	41	0	0	5
08-188	Fairy Lake, ON	43.621242	-80.048136	0.3	high	0.22	0.20	318	0	0	9
08-189	Schmidt Lake, ON	44.167604	-81.311053	0.3	moderate	0.31	0.01	4	0	0	32
08-191	Alder Lake, ON	43.353353	-80.537259	0.2	high	0.50	0.11	68	0	0	47
08-194	Irish Lake, ON	44.262603	-80.639939	0.3	moderate	0.18	0.21	18	0	0	12
08-195	Carson Lake, ON	44.621868	-81.25951	0.3	low	0.03	0.09	17	0	0	6
08-197	Copps Lake, ON	44.258274	-80.965301	0.1	low	0.11	0.01	3	0	0	10
08-201	Pike Lake, ON	43.967383	-80.816926	0.4	high	0.22	0.31	77	0	0	34
08-203	Arran Lake, ON	44.481854	-81.259812	4.0	moderate	0.23	0.03	6	0	0	50

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
08-204	Bartley Lake, ON	45.215178	-81.488534	0.1	low	0.00	0.02	0	0	0	0
08-207	Emmett Lake, ON	45.214271	-81.459651	1.0	low	0.00	0.00	0	0	0	0
08-209	Cameron Lake, ON	45.216979	-81.55796	1.6	low	0.00	0.02	1	0	0	0
08-213	Bass Lake, ON	45.889627	-81.937381	2.7	low	0.00	0.02	3	0	0	8
08-215	NA, ON	42.874465	-82.183136	0.1	high	0.73	0.11	103	1	0	22
08-216	Silver Lake, ON	45.878584	-82.891038	5.4	low	0.00	0.02	1	0	0	1
09-287	NA, SK	52.645297	-103.305742	0.4	high	0.69	0.04	1	0	0	7
09-288	NA, SK	54.285648	-104.640536	0.2	low	0.00	0.02	0	0	0	0
09-290	NA, SK	54.666389	-102.074369	0.3	moderate	0.00	0.23	93	0	0	0
09-296	Kaweenakumik Lake, MB	52.789186	-99.487028	73.7	low	0.00	0.00	0	0	0	0
09-298	Steepbank Lake, MB	50.903309	-98.035553	1.1	low	0.03	0.01	0	0	0	8
09-299	Husey Lake, MB	50.937946	-98.640674	1.6	low	0.01	0.02	1	1	1	10
09-300	Pine Lake, MB	50.88855	-98.31427	1.2	moderate	0.17	0.02	1	0	0	28
09-302	Norris Lake, MB	50.480156	-97.419123	0.5	low	0.02	0.05	6	0	0	4
09-367	Buck Lake, AB	52.985369	-114.762406	25.2	moderate	0.00	0.03	3	0	0	12
09-371	Millers Lake, AB	53.559871	-116.768414	0.4	moderate	0.02	0.25	38	0	0	3
09-372	NA, AB	53.682975	-116.270225	0.5	moderate	0.01	0.04	10	0	0	13
09-373	NA, AB	53.791313	-116.196712	2.1	low	0.01	0.01	1	0	0	2
09-374	NA, AB	53.560248	-114.320995	0.2	high	0.38	0.07	16	0	0	17
09-376	Peanut Lake, AB	54.017538	-114.349031	0.4	moderate	0.26	0.02	8	0	0	14

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
09-377	NA, AB	54.110058	-114.246698	0.2	high	0.67	0.03	7	0	0	29
09-378	Canoe Lake, AB	54.622591	-113.123462	1.0	high	0.51	0.03	3	0	0	15
09-379	Long Lake, AB	54.483	-112.754406	6.1	low	0.00	0.01	1	0	0	2
09-380	NA, AB	55.061393	-114.016717	0.8	moderate	0.00	0.01	1	0	0	7
09-381	Winagami Lake, AB	55.623088	-116.74359	45.1	moderate	0.15	0.01	0	0	0	1
09-382	Montagneuse Lake, AB	56.503692	-118.46527	0.4	low	0.00	0.01	0	0	0	0
09-383	Lac Magloire, AB	55.871434	-117.173848	7.7	high	0.75	0.02	1	0	0	2
09-384	Flyingshot Lake, AB	55.139873	-118.862224	1.4	high	0.51	0.13	26	1	0	14
09-391	Charron Lake, AB	54.847322	-112.508763	12.2	high	0.50	0.03	2	0	0	13
09-394	Elinor Lake, AB	54.646202	-111.652296	9.3	low	0.00	0.01	0	0	0	2
09-395	Upper Mann Lake, AB	54.143992	-111.499621	5.3	moderate	0.20	0.05	3	0	0	15
09-396	Vincent Lake, AB	54.116443	-111.341336	8.4	high	0.32	0.04	5	0	0	17
09-403	Wabamun Lake, AB	53.544071	-114.581865	77.3	moderate	0.03	0.08	7	0	0	8
09-408	Eleven Lake, MB	50.523187	-99.843826	0.3	moderate	0.24	0.03	1	0	0	12
09-410	NA, MB	50.572023	-100.190591	0.3	high	0.54	0.03	1	0	0	9
09-413	NA, MB	51.008286	-101.106274	0.3	moderate	0.14	0.14	2	0	0	2
09-418	Hazel Lake, SK	52.671132	-105.272507	1.0	high	0.29	0.02	1	0	0	2
09-419	NA, SK	52.722615	-105.209001	1.1	high	0.66	0.03	1	0	0	3
09-420	Jumping Lake, SK	52.855734	-105.447346	10.3	high	0.80	0.03	0	0	0	2

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
09-421	NA, SK	52.854199	-106.157499	1.0	high	0.63	0.03	5	0	0	12
09-423	NA, SK	53.183739	-106.844945	1.1	moderate	0.34	0.05	6	0	0	3
09-424	NA, SK	53.220024	-106.853005	1.2	moderate	0.11	0.02	3	0	0	3
09-425	NA, SK	53.165963	-106.814976	0.5	high	0.31	0.08	17	0	0	6
09-426	Sled Lake, SK	54.456237	-107.419508	91.5	low	0.00	0.00	0	0	0	0
09-429	Bisgrove Lake, MB	51.149512	-101.079876	0.9	high	0.48	0.02	1	0	0	14
09-431	Jeannette Lake, SK	54.541364	-108.547482	3.7	low	0.00	0.01	0	0	0	0
09-432	NA, SK	53.54476	-109.095614	0.2	high	0.52	0.05	6	0	0	18
09-447	Mistawasis Lake, SK	53.08939	-107.236605	6.3	low	0.05	0.01	0	0	0	3
09-451	Picnic Lake, SK	53.208341	-108.664809	0.5	moderate	0.19	0.02	1	0	0	7
09-454	Kerrs Lake, MB	50.496501	-99.688457	1.4	moderate	0.11	0.02	1	0	0	10
09-605	Swan Lake, BC	55.518181	-120.015159	5.5	low	0.01	0.01	1	0	0	2
09-606	Clairmont Lake, AB	55.256701	-118.76132	6.5	high	0.58	0.17	5	0	0	12
09-608	Cardinal Lake, AB	56.240918	-117.734668	48.6	moderate	0.27	0.02	1	0	0	5
10-276	Swan Lake, MB	49.360996	-98.911512	12.6	high	0.70	0.03	2	1	1	9
10-277	Gibsons Lake, MB	49.426958	-99.435161	0.2	low	0.02	0.01	0	0	0	0
10-278	Pelican Lake, MB	49.319927	-99.545126	27.5	high	0.73	0.03	1	1	0	11
10-280	Lake Stormon, MB	49.002982	-100.065083	0.3	low	0.00	0.04	1	0	0	0
10-281	Max Lake, MB	49.062751	-100.141199	2.7	low	0.00	0.03	0	0	0	0
10-285	Pasqua Lake, SK	50.785639	-103.952104	18.5	high	0.65	0.03	9	1	1	7
10-347	Outpost Lake, AB	49.011395	-113.456521	0.9	low	0.00	0.04	0	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
10-348	Shanks Lake, AB	49.068437	-112.718603	4.1	high	0.49	0.01	1	0	0	16
10-350	Keiver's Lake, AB	51.68812	-113.555192	1.1	high	0.68	0.04	2	0	0	26
10-351	Handhills Lake, AB	51.490556	-112.12657	8.7	moderate	0.40	0.02	1	0	0	29
10-353	Elkwater Lake, AB	49.667277	-110.295708	2.2	low	0.00	0.06	3	0	0	1
10-354	Gull Lake, SK	50.110293	-108.496956	0.6	moderate	0.27	0.06	1	0	0	10
10-360	Kenosee Lake, SK	49.813921	-102.309917	8.3	low	0.01	0.02	3	0	0	0
10-361	Gillis Lake, SK	49.839573	-102.425933	0.6	low	0.01	0.00	0	0	0	0
10-362	Pelican Lake, SK	50.542953	-106.012079	26.0	high	0.54	0.02	0	0	0	7
10-364	NA, SK	50.392956	-109.488101	0.3	high	0.79	0.02	0	0	0	7
10-365	Shooting Lake, AB	52.182962	-112.343561	5.0	moderate	0.36	0.02	0	0	0	18
10-386	NA, AB	53.463347	-113.228775	0.2	moderate	0.03	0.26	50	0	0	13
10-390	NA, AB	53.658603	-112.778849	0.0	low	0.00	0.01	0	0	0	0
10-401	NA, AB	52.471187	-113.172336	0.2	high	0.48	0.04	3	0	0	34
10-404	Hastings Lake, AB	53.417396	-112.911743	8.0	low	0.07	0.08	19	0	0	8
10-407	NA, SK	52.344099	-108.767665	0.1	high	0.75	0.04	6	0	0	3
10-414	Wilson Lake, SK	51.31598	-102.877558	0.3	high	0.84	0.01	0	0	0	8
10-415	Fishing Lake, SK	51.819993	-103.544821	30.2	high	0.77	0.03	1	0	0	3
10-417	Houghton Lake, SK	52.352834	-105.140944	15.3	high	0.75	0.03	1	0	0	4
10-422	Duck Lake, SK	52.802144	-106.263384	8.7	high	0.74	0.03	5	0	0	13
10-434	NA, SK	52.708506	-109.791664	1.4	moderate	0.41	0.02	3	0	0	16
10-436	Suffern Lake, SK	52.638124	-109.898419	0.3	low	0.00	0.03	0	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
10-437	Radisson Lake, SK	52.489493	-107.408292	4.4	high	0.69	0.02	1	0	0	4
10-439	Atton Lake, SK	52.84409	-108.859002	1.2	moderate	0.43	0.04	2	0	0	3
10-440	NA, SK	52.720884	-108.51576	0.7	moderate	0.51	0.02	0	0	0	4
10-441	NA, SK	52.980027	-107.810639	0.3	high	0.51	0.02	1	0	0	7
10-442	Tramping Lake, SK	52.003037	-108.784517	13.0	high	0.73	0.03	2	0	1	5
10-443	NA, SK	51.631497	-104.274473	0.3	low	0.20	0.06	1	0	0	1
10-446	Acton Lake, SK	52.916564	-107.873935	1.2	high	0.75	0.01	0	0	0	4
10-448	Redberry Lake, SK	52.689028	-107.159874	48.4	moderate	0.44	0.02	1	0	0	5
10-599	Bruce Lake, AB	52.382773	-110.969002	1.1	moderate	0.19	0.02	1	0	0	18
10-600	NA, AB	52.695158	-111.374896	0.7	moderate	0.23	0.05	0	0	0	5
10-601	NA, AB	53.547125	-111.801211	0.2	high	0.76	0.04	1	0	0	13
11-316	Slocan Lake, BC	49.932203	-117.393357	68.6	low	0.00	0.01	1	0	0	0
11-317	Victor Lake, BC	50.957539	-118.396542	0.1	low	0.00	0.01	0	0	0	0
11-343	Wasa Lake, BC	49.779692	-115.735377	1.0	moderate	0.00	0.12	13	0	0	0
11-344	Tie Lake, BC	49.410863	-115.327295	1.4	low	0.00	0.05	6	0	0	0
11-345	Surveyors Lake, BC	49.245773	-115.235662	0.1	moderate	0.00	0.21	6	0	0	0
11-471	Round Lake, BC	54.656922	-126.924746	1.7	moderate	0.03	0.02	3	0	0	27
11-476	Mulvaney Lake, BC	54.077005	-125.757675	0.3	moderate	0.00	0.05	11	0	0	5
11-477	Hanson Lake, BC	54.242465	-125.061656	1.7	low	0.00	0.00	0	0	0	0
11-479	NA, BC	53.42795	-124.545783	0.5	low	0.00	0.00	0	0	0	0
11-485	Eakin Lake, BC	53.810107	-125.913779	0.4	moderate	0.02	0.02	1	0	0	14

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
11-490	Eena Lake, BC	54.049179	-123.018163	0.5	moderate	0.00	0.16	6	0	0	2
11-492	Nukko Lake, BC	54.064509	-123.007832	4.2	moderate	0.02	0.04	3	0	0	4
11-494	Nulki Lake, BC	53.90613	-124.137247	15.7	low	0.02	0.00	0	0	0	3
11-496	Cummings Lake, BC	52.086749	-121.952157	0.5	moderate	0.00	0.00	1	0	0	16
11-497	NA, BC	52.000623	-122.173033	0.1	moderate	0.00	0.05	4	0	0	16
11-498	Chaunigan Lake, BC	51.571867	-123.893069	4.4	low	0.00	0.00	0	0	0	0
11-500	NA, BC	52.15161	-124.512345	0.4	low	0.00	0.00	0	0	0	0
11-501	Nimpo Lake, BC	52.340193	-125.204735	9.2	low	0.00	0.00	0	0	0	0
11-502	Blue Lake, BC	52.341465	-122.236095	0.4	low	0.01	0.01	0	0	0	4
11-510	Bridge Lake, BC	51.501772	-120.751359	13.4	moderate	0.00	0.03	1	0	0	4
11-511	Otter Lake, BC	51.530721	-120.678772	0.5	moderate	0.00	0.03	0	0	0	1
11-512	French Lake, BC	51.592882	-120.764438	0.6	moderate	0.00	0.00	1	0	0	0
11-513	Bouchie Lake, BC	53.035093	-122.626301	1.3	moderate	0.00	0.11	13	0	0	2
11-538	Rose Lake, BC	54.398593	-126.023267	0.2	moderate	0.00	0.12	8	0	0	6
11-539	Tachick Lake, BC	53.95755	-124.191784	20.6	moderate	0.03	0.01	1	1	0	5
11-540	NA, BC	54.574531	-123.807885	0.1	low	0.00	0.00	0	0	0	0
11-542	Witch Lake, BC	55.131569	-124.482396	15.1	low	0.00	0.00	0	0	0	0
11-543	NA, BC	55.334542	-125.318096	0.2	moderate	0.00	0.00	0	0	0	0
11-544	MacDonald Lake, BC	54.035583	-125.040308	0.2	moderate	0.00	0.14	2	0	0	0
11-545	Seas Lake, BC	54.051353	-124.777216	0.3	moderate	0.00	0.08	3	0	0	4

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
11-548	Mackenzie Lakes, BC	53.539827	-122.942903	0.3	low	0.00	0.01	0	0	0	0
11-554	Hammer Lake, BC	51.234899	-120.718194	0.6	low	0.00	0.00	0	0	0	0
11-558	Tabor Lake, BC	53.916375	-122.54217	3.7	moderate	0.00	0.14	16	0	0	1
11-590	Aleza Lake, BC	54.115384	-122.064291	0.6	low	0.00	0.00	0	0	0	0
11-595	Upper Little Slocan Lake, BC	49.67651	-117.658481	0.7	low	0.00	0.00	0	0	0	0
11-625	Crystal Lake, BC	54.416617	-122.618301	0.4	low	0.00	0.02	0	0	0	0
11-626	NA, BC	54.67022	-123.558782	0.2	low	0.00	0.00	0	0	0	0
11-631	Dragon Lake, BC	52.94939	-122.420822	5.3	moderate	0.01	0.16	44	0	0	24
11-632	Ahbau Lake, BC	53.268784	-122.097099	8.0	low	0.00	0.00	0	0	0	0
12-461	Burnaby Lake, BC	49.243327	-122.946538	0.3	high	0.00	0.92	3785	0	1	0
12-463	Garden Bay Lake, BC	49.647232	-124.02279	0.6	moderate	0.00	0.14	26	0	0	0
12-464	Lily Lake, BC	49.613256	-124.023197	0.1	moderate	0.00	0.23	77	0	0	0
12-465	Alta Lake, BC	50.114032	-122.981403	1.0	moderate	0.00	0.20	123	0	0	0
12-467	Green Lake, BC	50.151407	-122.931788	2.0	low	0.00	0.07	46	0	0	0
12-472	NA, BC	55.024461	-127.346246	0.4	low	0.00	0.00	1	0	0	0
12-474	Lakelse Lake, BC	54.381312	-128.559744	13.3	low	0.00	0.01	2	0	0	0
12-475	Diana Lake, BC	54.209495	-130.147927	2.5	low	0.00	0.01	0	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
12-532	Kitsumkalum Lake, BC	54.766909	-128.785448	18.4	low	0.00	0.00	0	0	0	0
12-536	Pine Lake, BC	54.614901	-128.7178	0.3	low	0.00	0.00	0	0	0	0
12-549	Laxton Lake, BC	49.008293	-122.35186	0.2	high	0.50	0.30	91	0	0	347
12-634	Elk Lake, BC	48.526599	-123.397351	2.2	high	0.03	0.47	164	0	0	21
12-635	Prospect Lake, BC	48.513468	-123.441651	0.7	moderate	0.00	0.20	76	0	0	1
12-638	Cusheon Lake, BC	48.815816	-123.468563	0.3	moderate	0.01	0.19	55	0	0	3
12-639	St Mary Lake, BC	48.890443	-123.542531	1.8	moderate	0.01	0.32	55	0	0	3
12-643	Glen Lake, BC	48.437398	-123.522473	0.2	high	0.00	0.40	595	0	0	0
12-645	Weeks Lake, BC	48.584406	-123.856961	0.3	low	0.00	0.00	0	0	0	0
12-647	Somenos Lake, BC	48.80178	-123.703764	1.0	high	0.03	0.30	186	0	1	26
12-650	Quennell Lake, BC	49.070025	-123.811737	1.1	high	0.05	0.28	88	0	0	17
12-652	Hoggan Lake, BC	49.151569	-123.8284	0.2	moderate	0.00	0.23	61	0	0	0
12-660	Nimpkish Lake, BC	50.419576	-126.978939	37.7	low	0.00	0.00	0	0	0	0
12-661	Roselle Lake, BC	50.520655	-126.989432	0.2	moderate	0.00	0.07	0	0	0	0
12-663	Alice Lake, BC	50.450374	-127.400686	10.8	low	0.00	0.00	0	0	0	0
12-664	Beaver Lake, BC	50.600735	-127.314969	0.2	moderate	0.00	0.08	0	0	0	0
12-666	Spirit Lake, BC	50.199099	-125.67783	0.3	moderate	0.00	0.00	0	0	0	0
12-667	Buttle Lake, BC	49.749851	-125.585696	37.1	low	0.00	0.00	0	0	0	0
12-668	Upper Quinsam Lake, BC	49.8808	-125.550404	5.0	low	0.00	0.00	0	0	0	0

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12-669	Echo Lake, BC	49.986046	-125.410997	0.2	low	0.00	0.02	0	0	0	0
12-673	Maple Lake, BC	49.636743	-125.01777	0.2	moderate	0.00	0.17	149	0	0	0
12-674	Chicadee Lake, BC	49.559465	-124.814538	0.2	low	0.01	0.07	7	0	0	3
12-678	Duck Lake, BC	49.85317	-124.445083	0.6	low	0.00	0.00	0	0	0	0
13-515	NA, BC	58.935329	-129.954645	0.1	moderate	0.00	0.08	0	0	0	0
13-518	Wye Lake, YT	60.063643	-128.699031	0.2	high	0.00	0.70	44	0	0	0
13-520	Pine Lake, YT	60.125379	-130.926778	0.2	low	0.00	0.01	0	0	0	0
13-522	Cliff Lake, BC	59.761821	-133.773316	0.1	moderate	0.00	0.03	1	0	0	0
13-524	Klukshu Lake, YT	60.322662	-137.015561	1.3	low	0.00	0.00	0	0	0	0
13-527	Enger Lakes, YT	62.256408	-140.66822	1.2	low	0.00	0.02	0	0	0	0
13-559	Kluachon Lake, BC	57.848343	-130.008819	1.1	low	0.00	0.01	3	0	0	0
13-561	Simmons Lake, BC	59.185497	-129.776374	0.5	low	0.00	0.00	0	0	0	0
13-564	Wolverine Lake, YT	61.455898	-130.240718	7.6	low	0.00	0.00	0	0	0	0
13-567	Little Salmon Lake, YT	62.187056	-134.685124	59.3	low	0.00	0.00	0	0	0	0
13-586	Fox Lake, YT	61.23237	-135.462882	15.2	low	0.00	0.00	0	0	0	0
13-588	NA, BC	59.979224	-127.55746	0.3	low	0.00	0.02	0	0	0	0
13-679	Kloo Lake, YT	60.958929	-137.862377	11.6	low	0.00	0.00	0	0	0	0
14-570	Gravel Lake, YT	63.809838	-137.894084	0.4	low	0.00	0.00	0	0	0	0
14-571	Chapman Lake, YT	64.848457	-138.347406	1.3	moderate	0.00	0.01	0	0	0	0
14-584	NA, YT	64.650174	-138.392056	0.2	low	0.00	0.00	0	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
17-012	Lac Fromenteau, QC	48.797033	-64.577585	0.9	low	0.00	0.00	4	0	0	0
17-017	Lac Baillargeon, QC	48.787719	-64.827227	0.9	moderate	0.00	0.00	0	0	0	0
17-018	Lac Bazire, QC	48.716045	-64.768398	0.2	low	0.00	0.00	0	0	0	0
17-027	Lac à l'Oie, QC	48.213122	-65.852432	0.3	moderate	0.01	0.00	5	0	0	6
17-034	Strachens Lake, NB	47.452333	-66.239988	0.1	moderate	0.00	0.02	0	0	0	0
17-041	Étang à la Truite, QC	48.690742	-66.767274	1.0	low	0.00	0.00	0	0	0	0
17-042	Serpentine Lake, NB	47.138484	-66.867145	5.5	moderate	0.00	0.01	0	0	0	0
17-044	Miramichi Lake, NB	46.460712	-66.972431	2.3	low	0.00	0.01	0	0	0	0
17-048	Lac Angus, QC	48.391954	-67.34011	0.3	low	0.00	0.01	3	0	0	0
17-053	Lac des Huit Milles, QC	48.408511	-67.577	0.7	moderate	0.08	0.01	19	0	0	5
17-058	Lac Antoine, QC	48.608711	-67.797322	0.1	high	0.23	0.02	2	0	0	24
17-060	Lac Michaud, QC	48.600798	-67.821122	0.4	high	0.15	0.04	6	0	0	22
17-061	Lac des Joncs, QC	48.359371	-68.153954	0.5	high	0.15	0.01	5	0	0	30
17-062	Lac Aubin, QC	48.557429	-68.16172	0.1	high	0.04	0.00	10	0	0	12
17-063	Lac Neigette, QC	48.289761	-68.406804	0.7	high	0.09	0.01	9	0	0	17
17-064	Lac Plourde, QC	48.290929	-68.452249	0.2	high	0.10	0.05	39	1	0	26
17-065	Grand lac Squatec, QC	47.67184	-68.568464	12.4	low	0.00	0.00	1	1	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
17-067	Grand lac Touradi, QC	48.101149	-68.691699	7.7	low	0.00	0.00	0	0	0	0
17-068	Lac Baker, NB	47.370377	-68.700947	5.9	moderate	0.00	0.03	9	0	0	12
17-073	Lac de la Station, QC	48.281042	-68.874287	0.6	moderate	0.03	0.01	6	0	0	6
17-074	Lac des Îles, QC	48.110804	-68.883295	0.6	low	0.00	0.00	0	0	0	0
17-077	Lac Bertrand, QC	47.946902	-69.225446	0.1	moderate	0.05	0.03	5	0	0	12
17-078	Lac Rond, QC	47.764893	-69.329789	0.7	low	0.00	0.00	1	0	0	0
17-082	Lac Saint-Pierre, QC	47.413793	-69.895023	0.6	moderate	0.00	0.02	15	0	0	3
17-086	Lac de la Dame, QC	45.891603	-70.360266	0.1	low	0.00	0.00	0	0	0	0
17-089	Lac Chartier, QC	46.818612	-70.508205	0.3	low	0.00	0.00	1	0	0	0
17-091	Lac Fortin, QC	46.117487	-70.857676	1.6	moderate	0.00	0.03	17	0	0	29
17-099	Lac Lindsay, QC	45.180669	-71.542226	0.6	moderate	0.02	0.01	8	0	0	20
17-106	Les Trois Lacs, QC	45.800588	-71.889269	2.5	high	0.07	0.01	8	1	0	18
17-108	Lac Massawippi, QC	45.220903	-72.00096	19.3	high	0.09	0.03	16	1	0	25
17-112	Lac des Français, QC	45.441569	-72.224891	0.2	moderate	0.01	0.02	21	0	0	4
17-113	Spooner Pond, QC	45.738752	-72.144113	0.3	high	0.13	0.01	11	0	0	19
17-116	Lac Brais, QC	45.456527	-72.206288	0.5	moderate	0.00	0.01	8	0	0	8
17-121	Lac Waterloo, QC	45.334019	-72.519167	1.4	high	0.00	0.17	115	0	0	8
17-122	Lac Roxton, QC	45.467017	-72.653542	2.0	high	0.03	0.10	88	0	0	26

Lake ID	Lake name	Latitude	Longitude	Lake size (km²)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
18-318	Little White Lake, BC	50.876922	-119.313477	0.4	moderate	0.00	0.02	6	0	0	0
18-319	Gardom Lake, BC	50.603381	-119.200222	0.8	moderate	0.00	0.11	16	0	0	4
18-320	Otter Lake, BC	50.411597	-119.250063	0.6	high	0.05	0.09	44	0	0	39
18-321	Pillar Lake, BC	50.5935	-119.639506	0.4	low	0.00	0.02	3	0	0	0
18-322	Bolean Lake, BC	50.537209	-119.500897	0.7	moderate	0.00	0.01	0	0	0	0
18-323	Swan Lake, BC	50.318415	-119.256066	4.1	high	0.08	0.24	95	0	0	24
18-325	Swalwell Lake, BC	50.04874	-119.239608	2.9	low	0.00	0.01	0	0	0	0
18-326	Wood Lake, BC	50.081734	-119.389801	9.1	high	0.04	0.10	57	1	0	4
18-327	Shannon Lake, BC	49.85686	-119.612246	0.1	high	0.00	0.32	297	0	0	0
18-328	Upper Buse Lake, BC	50.621071	-120.049744	0.1	moderate	0.00	0.01	1	0	0	5
18-329	Roche Lake, BC	50.472222	-120.152106	1.6	low	0.00	0.00	0	0	0	0
18-330	Scuitto Lake, BC	50.544062	-120.140745	0.9	low	0.00	0.00	0	0	0	7
18-331	Douglas Lake, BC	50.149715	-120.237859	6.5	moderate	0.00	0.00	0	0	0	4
18-332	Kentucky Lake, BC	49.89656	-120.564275	0.4	low	0.00	0.02	0	0	0	0
18-333	Paradise Lake, BC	49.915249	-120.276864	1.2	moderate	0.00	0.01	0	0	0	0
18-334	Elkhart Lake, BC	49.894727	-120.30996	0.2	low	0.00	0.01	0	0	0	0
18-340	Mahoney Lake, BC	49.289105	-119.582061	0.2	low	0.00	0.02	2	0	0	0
18-341	Silver Lake, BC	49.82899	-119.83933	0.1	low	0.00	0.00	0	0	0	0
18-468	Anderson Lake, BC	50.643861	-122.401882	29.0	low	0.00	0.00	1	0	0	0

Lake ID	Lake name	Latitude	Longitude	Lake size (km^2)	Human impact class	Agricultural fraction	Urban fraction	Population density	WWTPs presence	Hospitals presence	Livestock density
18-469	Gun Lake, BC	50.872838	-122.879031	5.7	moderate	0.00	0.04	2	0	0	0
18-506	Whitewood Lake, BC	51.097401	-120.34108	0.2	low	0.00	0.00	0	0	0	0
18-507	Dutch Lake, BC	51.651898	-120.055866	0.5	high	0.00	0.33	151	0	0	0
18-550	Pimainus Lakes, BC	50.403098	-121.085132	0.6	moderate	0.00	0.00	0	0	0	0
18-551	Shambrook Lake, BC	50.485507	-120.50673	0.1	moderate	0.00	0.06	1	0	0	0
18-555	Cultus Lake, BC	50.859036	-121.053029	0.4	low	0.00	0.01	0	0	0	0
18-591	Allison Lake, BC	49.698817	-120.605845	0.6	low	0.00	0.01	1	0	0	1
18-592	Missezula Lake, BC	49.789563	-120.525908	2.5	low	0.00	0.01	0	0	0	0
18-594	Ellison Lake, BC	49.992999	-119.394635	2.0	moderate	0.01	0.02	6	1	0	1

A2.1. R script for the negative binomial model

```
library(tidyverse)
library(MASS)
library(DHARMa)
library(MKpower)
library(quantreg)
library(Hmisc)
library(rms)
library(PResiduals)
library(lubridate)

setwd("WorkingDirectory/BinomialModel")

# 1 Explanatory variables----
data1 <- read.csv("explanatoryVariables.csv")
data1$res_time[data1$res_time==-1]<-45500
data1 <- data1[,c(1,3,4,9,10,11,12,20,25,37,38)]
data2 <- read.csv("pop_density.csv")
data3 <- merge(data1, data2, by="lakeID")

# 2 Outcomes----
data4 <- read.csv("responseVariables.csv")
data4 <- data4[, c(1,2)]

# 3 Merge- Final data----
data <- merge(data3, data4, by="lakeID")

# 4 Data format----
data$sampling_date <- as.Date(data$sampling_date, format = "%m/%d/%Y")

data$area_ratio <- as.numeric(data$area_ratio)
data$ln_area_ratio <- as.numeric(log(as.numeric(data$area_ratio)))

data$lake_depth <- as.numeric(data$lake_depth)
data$ln_lake_depth <- as.numeric(log(as.numeric(data$lake_depth)))

data$res_time <- as.numeric(data$res_time)

data$ln_res_time <- as.numeric(log(as.numeric(data$res_time)))

data$mean_slope <-as.numeric(data$mean_slope)
```

```
data$ln_mean_slope <-as.numeric(log(as.numeric(data$mean_slope)))

data$prec <-as.numeric(as.numeric(data$precipitationSum7daysPrior)*100)

data$urb_frac <-as.numeric(data$urban_frac)
data$urb_frac_per <-as.numeric(data$urban_frac*100)

data$agr_frac <-as.numeric(data$agriculture_frac)
data$agr_frac_per <-as.numeric(data$agr_frac*100)

data$wwtps_pres <-as.factor(data$wwtps_pres)

data$pop_density <- as.numeric(data$pop_density)

data$nb_overall <-as.integer(data$nb_overall)

# * 4.1 Changing date to days from jan 1----
data$days <-yday(data$sampling_date)-1
data$days <- as.numeric(data$days)

# 5 Histograms----
par(mfrow=c(1,2))
hist(data$urb_frac)
hist(data$agr_frac)
hist(data$pop_density)
hist(data$area_ratio)
hist(data$ln_area_ratio)
hist(data$lake_depth)
hist(data$ln_lake_depth)
hist(data$res_time)
hist(data$ln_res_time)
hist(data$mean_slope)
hist(data$ln_mean_slope)
hist(data$prec)
hist(data$days)
table(data$wwtps_pres)

# 6 Observations missing res_time are out (n=6)- Final data----
data<-data[!is.na(data$res_time), ]

# 7 Lakes that may be outliers----
data <- data[data$lakeID != "07-231",]
```

```
# 8 Negative Binomial model----
model.nb <- glm.nb(nb_overall ~ agr_frac_per + urb_frac_per + wwtps_pres + area_ratio +
                   ln_lake_depth + ln_res_time + ln_mean_slope + prec + days, data=data)

summary(model.nb)
exp(coef(model.nb))
exp(confint(model.nb))
vif(model.nb)

# 9 Diagnostics of Goodness of Fit (Residual Analysis)----
# * 9.1Simulated residuals----
# https://cran.r-project.org/web/packages/DHARMa/vignettes/DHARMa.html

set.seed(123)
simulationOutput <- simulateResiduals(fittedModel = model.nb, plot = T)
testResiduals(simulationOutput)
```

A2.2. R script for the ordinal logistic regression model for total TrOCs' sum

```
library(MKpower)
library(quantreg)
library(tidyverse)
library(Hmisc)
library(ggplot2)
library(rms)
library(PResiduals)
library(MASS)
library(sure)
library(lubridate)

setwd("WorkingDirectory/OrdinalLogisticRegressionModel")

# 1 Explanatory variables----

data1 <- read.csv("explanatoryVariables.csv")

data1$res_time[data1$res_time==-1]<-45500

data1 <- data1[,c(1,3,4,9,10,11,12,20,25,37,38)]

data2 <- read.csv("pop_density.csv")
```

```
data3 <- merge(data1, data2, by="lakeID")

# 2 Outcomes----

data4 <- read.csv("responseVariables.csv")
data4 <- data4[, c(1,3)]

# 3 Merge- Final data----

data <- merge(data3, data4, by="lakeID")

# 4 Data format----
data$sampling_date <- as.Date(data$sampling_date, format = "%m/%d/%Y")

data$area_ratio <- as.numeric(data$area_ratio)
data$ln_area_ratio <- as.numeric(log(as.numeric(data$area_ratio)))

data$lake_depth <- as.numeric(data$lake_depth)
data$ln_lake_depth <- as.numeric(log(as.numeric(data$lake_depth)))

data$res_time <- as.numeric(data$res_time)
data$ln_res_time <- as.numeric(log(as.numeric(data$res_time)))

data$mean_slope <-as.numeric(data$mean_slope)
data$ln_mean_slope <-as.numeric(log(as.numeric(data$mean_slope)))

data$prec <-as.numeric(as.numeric(data$precipitationSum7daysPrior)*100)

data$urb_frac <-as.numeric(data$urban_frac)
data$urb_frac_per <-as.numeric(data$urban_frac*100)

data$agr_frac <-as.numeric(data$agriculture_frac)
data$agr_frac_per <-as.numeric(data$agr_frac*100)

data$wwtps_pres <-as.factor(data$wwtps_pres)

data$pop_density <- as.numeric(data$pop_density)

data$sum_overall <-as.numeric(data$sum_overall)

# 5 Changing date to days from jan 1----
data$days <-yday(data$sampling_date)-1
```

```
data$days <- as.numeric(data$days)

# 6 Observations missing res_time are out (n=6) - Final data----
data<-data[!is.na(data$res_time), ]

# 7 Ordinal Logistic Regression model----
orm.logit <- orm(sum_overall ~ agr_frac_per + urb_frac_per  + wwtps_pres  + area_ratio +
                    ln_lake_depth+ln_res_time +  ln_mean_slope + prec + days, data = data,
                    x=TRUE, y=TRUE)
orm.logit

x <- cbind(orm.logit$coefficients)

# 8 Residuals----
presid.logit <- presid (orm.logit)
plot(data$agr_frac_per, presid.logit)
plot(data$urb_frac_per, presid.logit)
plot(data$area_ratio, presid.logit)
plot(data$ln_lake_depth, presid.logit)
plot(data$ln_res_time, presid.logit)
plot(data$ln_mean_slope, presid.logit)
plot(data$prec, presid.logit)
plot(data$days, presid.logit)
plot(data$wwtps_pres, presid.logit, xlab="wwtps")
legend("topright",legend = c("0=No", "1=Yes"),pch = 19)
```

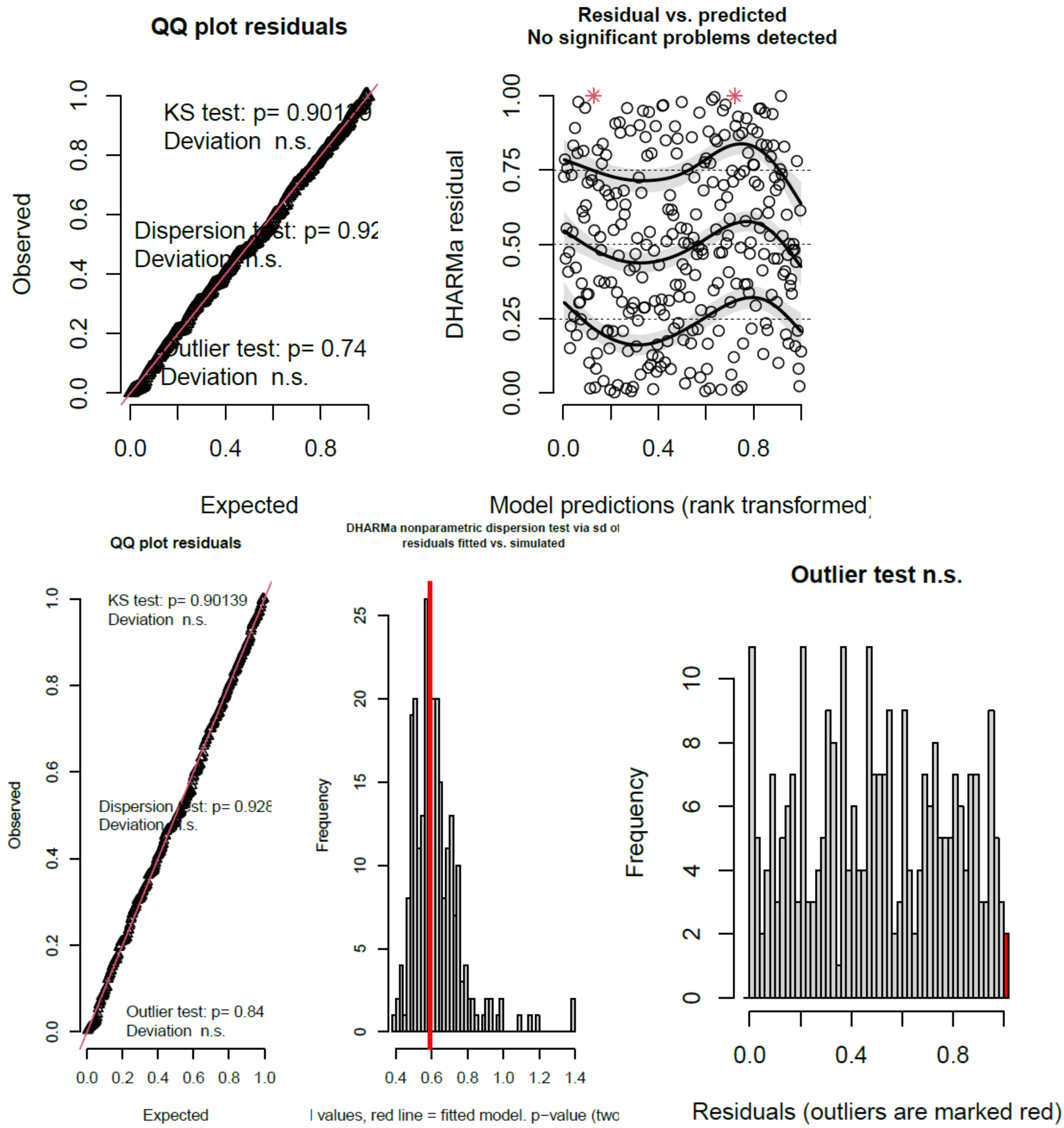

Figure 13. Model fit validation for negative binomial model.

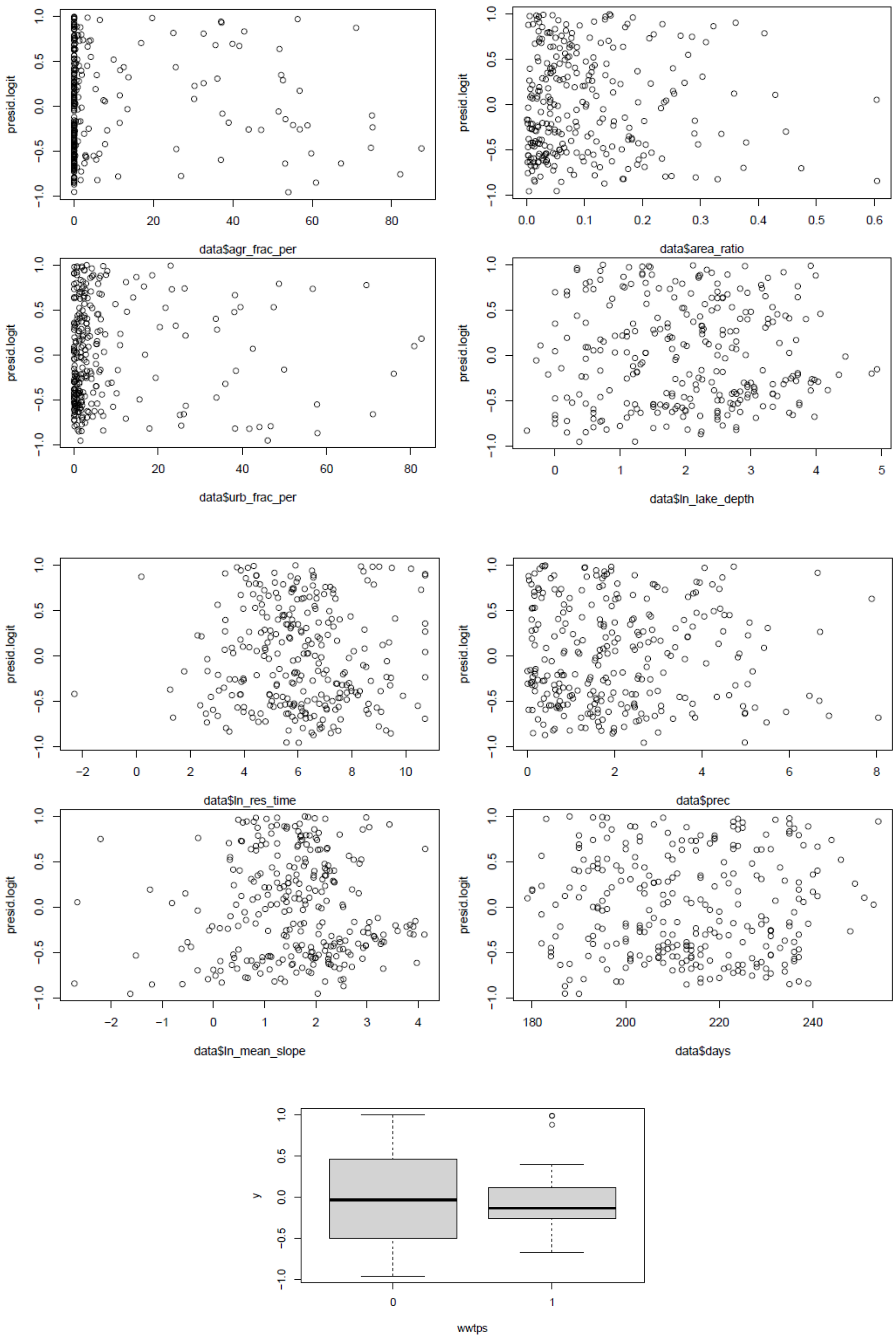

Figure 14. Model fit validation for ordinal logistic regression model for total TrOCs' sum.

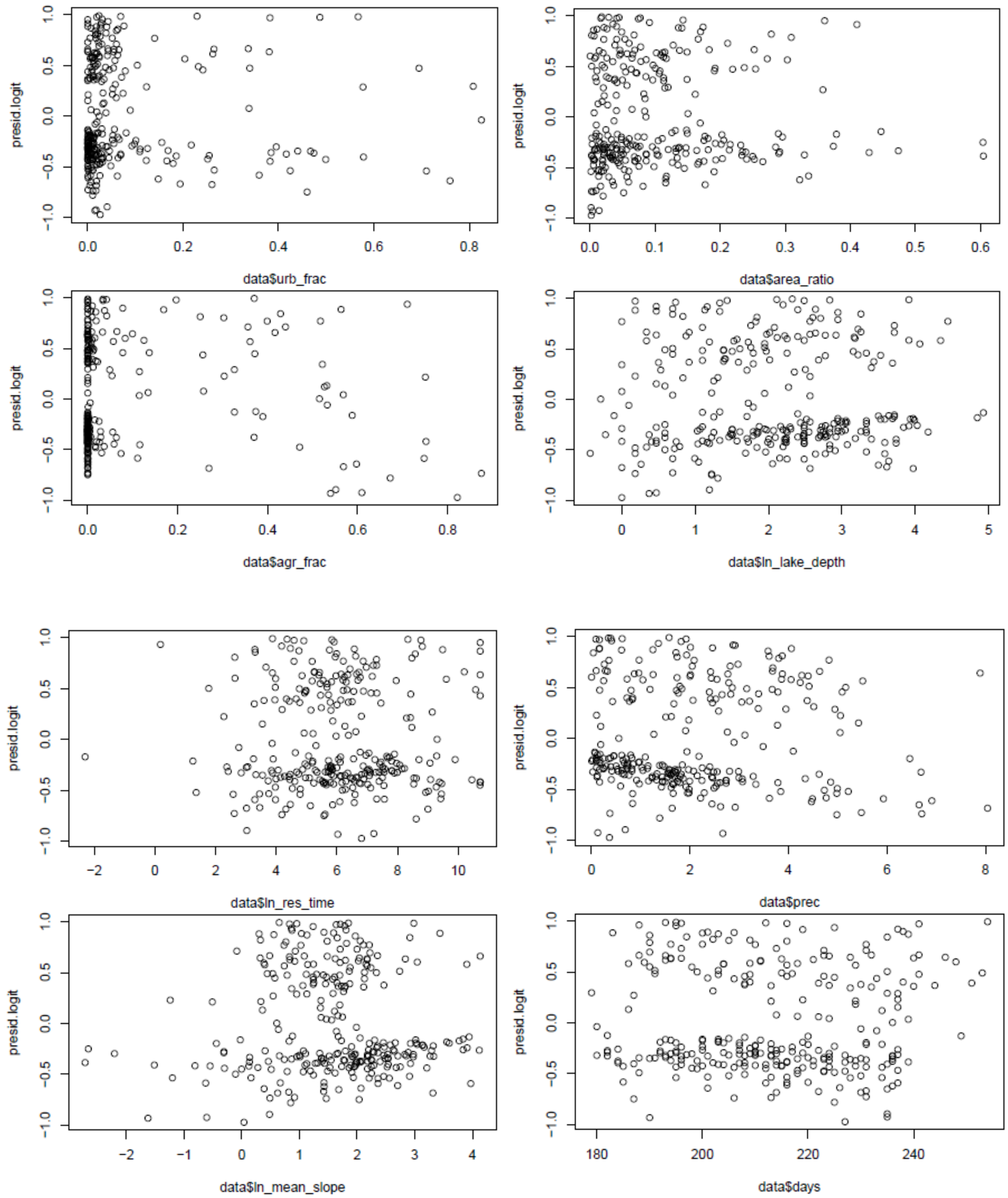

Figure 15. Model fit validation for ordinal logistic regression model for pesticides' sum.

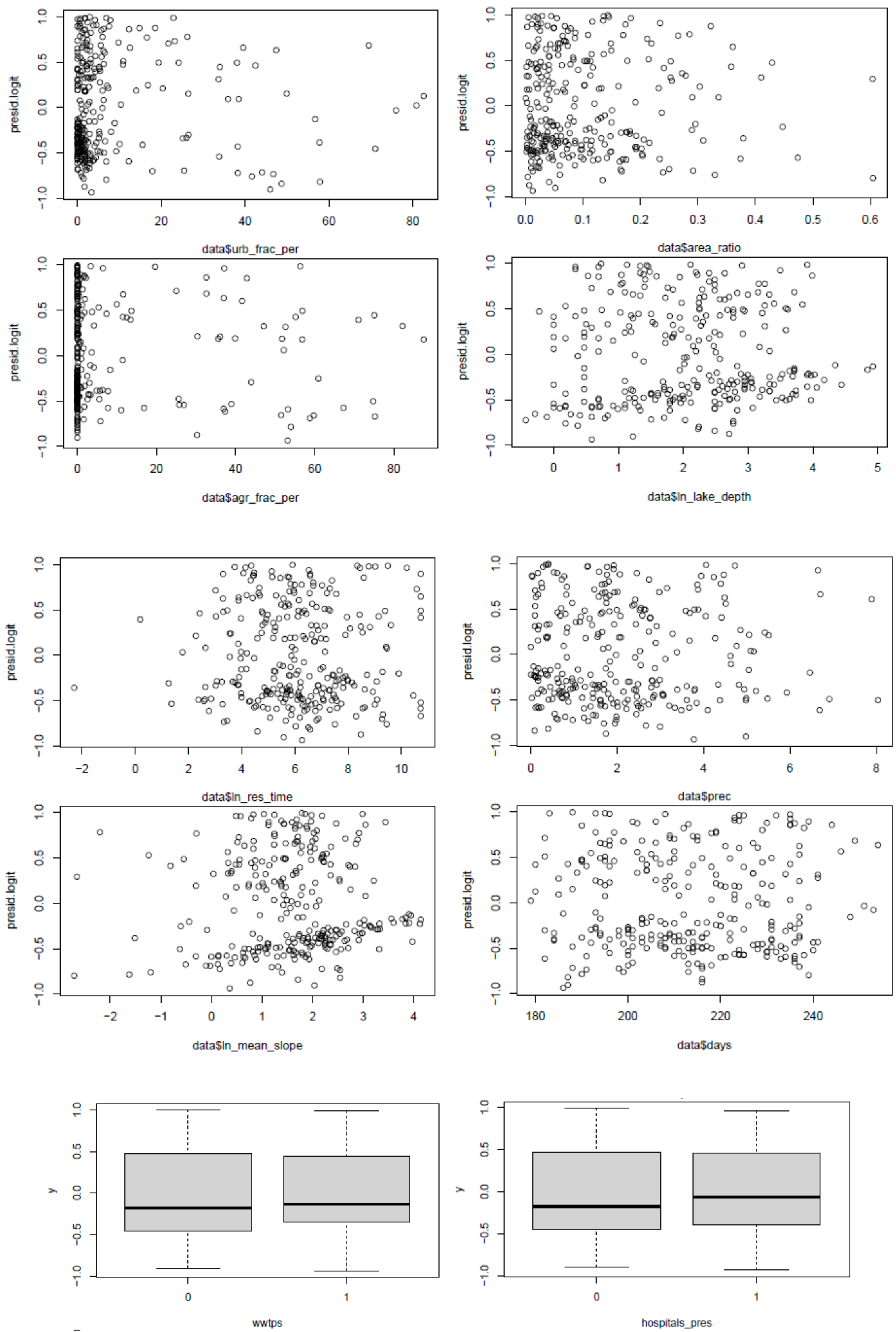

Figure 16. Model fit validation for ordinal logistic regression model for PPCPs and additives' sum.

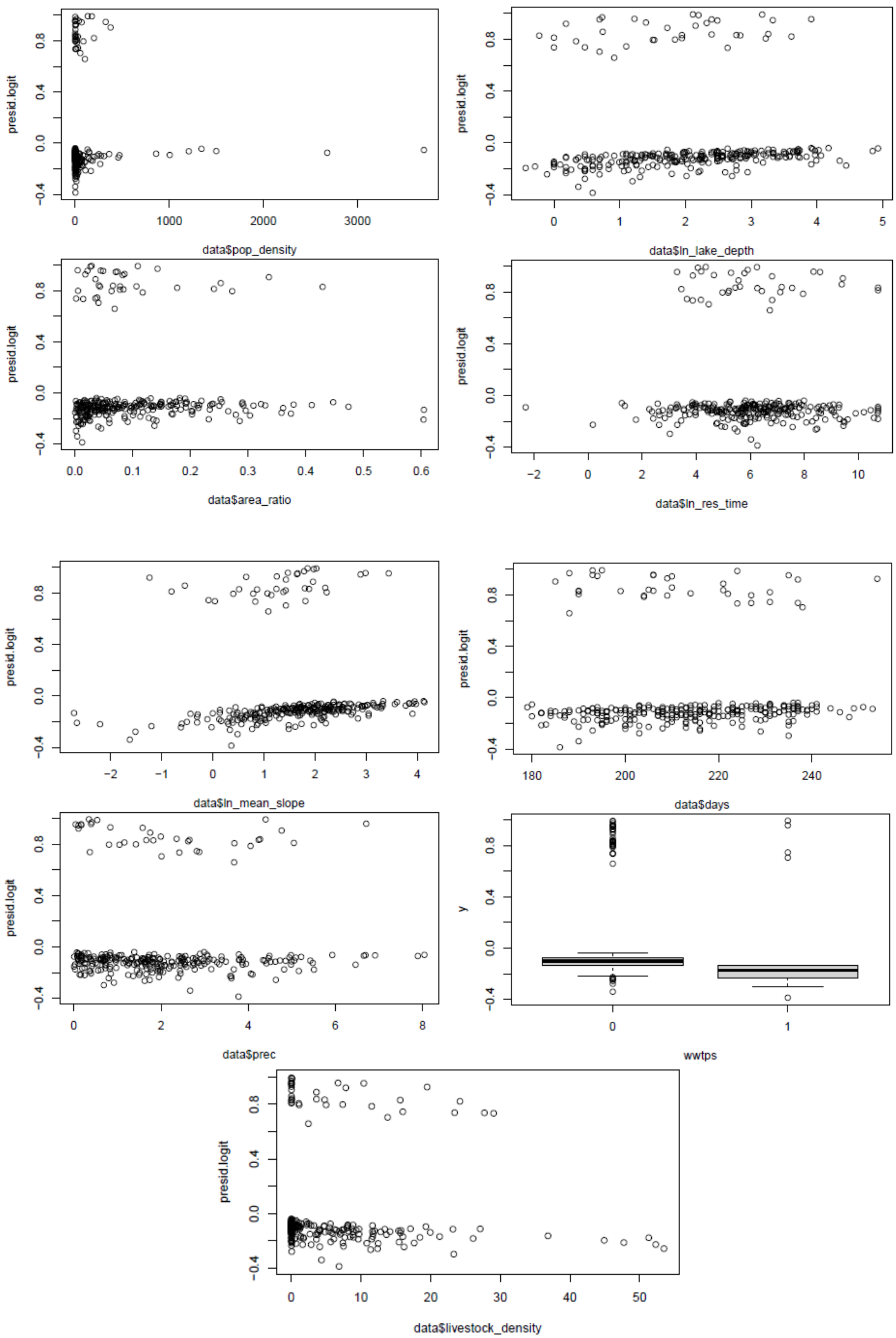

Figure 17. Model fit validation for ordinal logistic regression model for antibiotics' sum.