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Occurrence and Reductions of Pharmaceuticals and Personal Care Products and Estrogens by Municipal Wastewater Treatment Plants in Ontario, Canada

BY:

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ABSTRACT

Over the last ten years there have been reports of pharmaceuticals and personal care product (PPCP) residuals in municipal wastewater treatment plant (WWTP) effluents. The goals of this study were: (1) to expand the Canadian database of municipal wastewater influent and effluent concentrations for selected PPCPs; (2) to determine if three treatment configurations (e.g. lagoons, conventional activated sludge (CAS), and CAS followed by media filtration (CAS + filtration)) affect PPCP reduction; (3) to explore whether CAS operational parameters were correlated with reduction. Selected PPCPs included ten acidic pharmaceuticals, triclosan, five polycyclic musks and two estrogens. The pharmaceuticals and musks were selected on the basis of levels of use in Canada; reported aquatic toxicity effects; and the ability to analyze for the compounds at low levels. Twelve WWTPs discharging into the Thames River, the second largest river in southwestern Ontario, were surveyed.

Acidic pharmaceuticals were detected in all WWTP influent samples. The most frequently detected compounds in the influent samples were ibuprofen (IBU), gemfibrozil (GMF), naproxen (NPX) and diclofenac (DCF) (13.7 µg/L, 0.453 µg/L, 5.58 µg/L, and 0.204 µg/L respectively), whereas clofibric acid, fenoprofen, and fenofibrate were not detected in any influent samples. Reduction was consistently high (86-99%) for some compounds (i.e. IBU and NPX) while reduction of ketoprofen (KTP), DCF and Indomethacin (IND) was lower and highly variable. Lagoon systems had enhanced reductions. Triclosan (TCL) was detected in influent samples at concentrations of 4.01 µg/L to 0.01 µg/L and 0.01 to 0.324 µg/L in effluent samples. Reduction of TCL ranged from 74 – 95%; lagoon systems generated an effluent with below-measurable TCL concentrations.

The most frequently detected polycyclic musks were Galaxolide (HHCB) and Tonalide (AHTN). Median influent concentrations of HHCB and AHTN were 5.20 µg/L and 2.00 µg/L respectively. Median effluent concentrations for HHCB and AHTN were 1.82 µg/L and 0.600 µg/L respectively. Lagoon treatment systems had higher reductions than the CAS or CAS+filtration systems.

The hormones 17-β-estradiol (E2) and estrone (E1) were detected at concentrations of 0.006 µg/L to 0.014 µg/L and 0.016 µg/L to 0.049 µg/L respectively. E2 was not detected in any effluent samples (<0.005 µg/L) whereas E1 was detected in effluent samples from CAS treatment plants (median of 0.008 µg/L), and in one sample from lagoons.

These data demonstrate that there are detectable levels of PPCPs entering Canadian waterways at trace levels, and that only some of these compounds are being reduced in a significant proportion by municipal wastewater treatment processes.

NWRI RESEARCH SUMMARY

Plain language title

Occurrence and Reductions of Pharmaceuticals and Personal Care Products by Wastewater Treatment Plants in Ontario, Canada

What is the problem and what do scientists already know about it?

Pharmaceuticals and Personal Care Products (PPCP) are present in the effuents of sewage treatment plants. This study quantifies effluent concentrations of "acidic pharmaceuticals" and musk compounds. Influent concentrations were also characterized to determine if the level of treatment and the operating conditions of the treatment system had an impact on the removal of PPCP compounds. PPCPs are emerging contaminants. Analytical methods used in this study were developed by NWRI Research Scientist. The Canadian database for releases of PPCPs by sewage treatment plants is very limited.

Why did NWRI do this study?

NWRI did this study to establish a database that could be used for effects work and policy decisions.

What were the results?

For the following PPCPs: ten acidic pharmaceuticals, triclosan, five polycyclic musks and two hormones concentration data are given. Removal values of selected PPCPS by the twelve treament plants are presented.

How will these results be used?

Effects work. Policy formation.

Who were our main partners in the study?

Ontario Ministry of Environment, Great Lakes Sustainability Fund (Burlington, Ontario), Wastewater Technology Center (Burlington, Ontario)

Présence et réduction des résidus de produits pharmaceutiques, produits de soins personnels et œstrogènes par les installations municipales de traitement des eaux usées en Ontario (Canada)

L. Lishman, S.A. Smyth, Kurtis Sarafin, S. Kleywegt, J. Toito, T. Peart, B. Lee, M. Servos, M. Beland et P. Seto

RÉSUMÉ

Au cours des dix dernières années, des résidus de produits pharmaceutiques et produits de soins personnels (PPSP) ont été trouvés dans les effluents des installations municipales de traitement des eaux usées (ITEU). Voici les objectifs de cette étude : (1) développer la base de données canadienne relative aux concentrations d'influents et d'effluents pour certains PPSP; (2) déterminer si trois types de traitement (par exemple, lagunes, boues activées classiques (BAC) et procédé BAC suivi d'une filtration [BAC + filtration]) ont un effet sur la réduction des PPSP; (3) étudier si les paramètres opérationnels du procédé BAC ont un rapport avec cette réduction. Les PPSP retenus comprenaient dix produits pharmaceutiques acides, du triclosan, cinq muscs polycycliques et deux œstrogènes. Les produits pharmaceutiques et les muscs ont été sélectionnés en fonction de leur taux d'utilisation au Canada, de leur toxicité pour les organismes aquatiques et de la facilité de dosage de ces composés à de faibles concentrations. Douze ITEU se déchargeant dans la rivière Thames, la deuxième plus grande rivière du sud-ouest de l'Ontario, ont été étudiées.

Des produits pharmaceutiques acides ont été découverts dans les échantillons d'influent de toutes les ITEU. Les composés les plus fréquemment trouvés dans les échantillons d'influent ont été l'ibuprofène (IBU), le gemfibrozil (GMF), le naproxène (NPX) et le diclofénac (DCF) (13,7 µg/L, 0,453 µg/L, 5,58 µg/L et 0,204 µg/L respectivement), tandis que l'acide clofibrique, le fenoprofène et le fénofibrate n'ont été détectés dans aucun échantillon d'influent. La réduction obtenue était régulièrement forte (de 86 à 99 %) pour certains composés (comme l'IBU et le NPX), tandis que celle du ketoprofène (KTP), du DCF et de l'indométacine (IND) était faible et très variable. Les procédés de lagunage amélioraient les taux de réduction. Le triclosan (TCL) a été détecté à des concentrations de 4,01 µg/L à 0,01 µg/L dans les échantillons d'influent, et de 0,01 à 0,324 µg/L dans les échantillons d'effluent. Une réduction de TCL de 74 à 95 % a été mise en évidence; les procédés de lagunage ont produit un effluent dans lequel les concentrations de TCL n'étaient pas mesurables.

Les muscs polycycliques les plus fréquemment trouvés ont été le galaxolide (HHCB) et la tonalide (AHTN). Les concentrations médianes de HHCB et d'AHTN dans les influents étaient de 5,20 µg/L et de 2,00 µg/L respectivement. Les concentrations médianes de HHCB et d'AHTN dans les effluents étaient de 1,82 µg/L et de 0,600 µg/L respectivement. Les procédés de traitement par lagunage ont obtenu de meilleures réductions que les procédés de BAC ou de BAC + filtration.

Les hormones 17-β-estradiol (E2) et estrone (E1) étaient présentes à des concentrations de 0,006 µg/L à 0,014 µg/L et de 0,016 µg/L à 0,049 µg/L respectivement. L'hormone E2 n'a été trouvée dans aucun échantillon d'effluent (< 0,005 µg/L) tandis que l'E1 a été détectée dans les échantillons d'effluent des installations de traitement par BAC (concentration médiane de 0,008 µg/L) et dans un seul échantillon d'effluent des procédés de lagunage.

Ces données indiquent que des quantités détectables de PPSP pénètrent dans les eaux canadiennes à des concentrations traces, et que seuls certains composés sont réduits dans des proportions significatives par les procédés municipaux de traitement des eaux usées.

Sommaire des recherches de l'INRE

Titre en langage clair

Présence et réduction des résidus de produits pharmaceutiques et produits de soins personnels par les installations de traitement des eaux usées en Ontario (Canada)

Quel est le problème et que savent les chercheurs à ce sujet?

Des produits pharmaceutiques et produits de soins personnels (PPSP) sont présents dans les effluents des installations de traitement des eaux usées. Cette étude quantifie les concentrations de composés de muscs et de produits pharmaceutiques acides dans les effluents. On a aussi mesuré les concentrations dans les influents afin de déterminer si le taux de traitement et les conditions d'exploitation du procédé de traitement ont un impact sur l'élimination des PPSP. Les PPSP représentent de nouveaux contaminants. Les méthodes d'analyse utilisées dans cette étude ont été élaborées par des chercheurs de l'INRE. La base de données canadienne concernant les rejets de PPSP par les installations de traitement des eaux usées est très limitée.

Pourquoi l'INRE a-t-il effectué cette étude?

L'INRE a réalisé cette étude afin d'élaborer une base de données qui pourrait être utilisée pour la prise de décisions stratégiques et pour des travaux relatifs aux effets sur l'environnement.

Quels sont les résultats?

Pour les PPSP suivants : dix produits pharmaceutiques acides, le triclosan, cinq muscs polycycliques et deux hormones, les concentrations sont fournies. Le rapport présente les taux d'élimination des PPSP sélectionnés pour les douze installations de traitement.

Comment ces résultats seront-ils utilisés?

Travaux relatifs aux effets sur l'environnement. Élaboration des politiques.

Quels étaient nos principaux partenaires dans cette étude?

Ministère de l'Environnement de l'Ontario, Fonds de durabilité des Grands Lacs (Burlington, Ontario), Centre de technologie des eaux usées (Burlington, Ontario)

Occurrence and Reductions of Pharmaceuticals and Personal Care Products and Estrogens by Municipal Wastewater Treatment Plants in Ontario, Canada.

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ABSTRACT

Over the last ten years there have been reports of pharmaceuticals and personal care product (PPCP) residuals in municipal wastewater treatment plant (WWTP) effluents. The goals of this study were: (1) to expand the Canadian database of municipal wastewater influent and effluent concentrations for selected PPCPs; (2) to determine if three treatment configurations (e.g. lagoons, conventional activated sludge (CAS), and CAS followed by media filtration (CAS + filtration)) affect PPCP reduction; (3) to explore whether CAS operational parameters were correlated with reduction. Selected PPCPs included ten acidic pharmaceuticals, triclosan, five polycyclic musks and two estrogens. The pharmaceuticals and musks were selected on the basis of levels of use in Canada; reported aquatic toxicity effects; and the ability to analyze for the compounds at low levels. Twelve WWTPs discharging into the Thames River, the second largest river in southwestern Ontario, were surveyed.

Acidic pharmaceuticals were detected in all WWTP influent samples. The most frequently detected compounds in the influent samples were ibuprofen (IBU), gemfibrozil (GMF), naproxen (NPX) and diclofenac (DCF) (median concentrations of 13.7 μg/L, 0.453 μg/L, 5.58 μg/L, and 0.204 μg/L respectively), whereas clofibric acid, fenoprofen, and fenofibrate were not detected in any influent samples. Reduction was consistently high (86-99%) for some compounds (i.e. IBU and NPX) while reduction of ketoprofen (KTP), DCF and indomethacin (IND) was lower and highly variable. Lagoon systems had enhanced reductions. Triclosan (TCL) was detected at concentrations of 4.01 μg/L to 0.01 μg/L in influent samples and 0.01 to 0.324 μg/L in effluent samples. Reduction of TCL ranged from 74 – 95%; lagoon systems generated an effluent with below-measurable TCL concentrations.

The most frequently detected polycyclic musks were Galaxolide (HHCB) and Tonalide (AHTN). Median influent concentrations of HHCB and AHTN were 5.20 µg/L and 2.00 µg/L respectively. Median effluent concentrations for HHCB and AHTN were 1.82 µg/L and 0.600 µg/L respectively. Lagoon treatment systems had higher reductions than the CAS or the CAS+filtration systems.

The hormones 17-β-estradiol (E2) and estrone (E1) were detected at concentrations of 0.006 µg/L to 0.014 µg/L and 0.016 µg/L to 0.049 µg/L respectively. E2 was not detected in any effluent samples (<0.005 µg/L) whereas E1 was detected in effluent samples from CAS treatment plants (median of 0.008 µg/L), and in one sample from lagoons.

These data demonstrate that there are detectable levels of PPCPs entering Canadian waterways at trace levels, and that only some of these compounds are being reduced in a significant proportion by municipal wastewater treatment processes.

Keywords: municipal wastewater, pharmaceuticals, musks, estrogens

1. Introduction

In recent years, the fate and effects of pharmaceuticals and personal care products (PPCPs) entering the environment has gained increasing attention. PPCPs represent a large group of compounds which include non-prescription and prescription pharmaceuticals for human and veterinary use, and the active and inert ingredients in personal care products (Daughton and Ternes, 1999). Examples of PPCPs include analgesics, lipid regulators, natural and synthetic hormones, steroids, fragrances, sun screens, shampoos and cosmetics.

Most PPCPs, in their original or biologically altered form, are discharged into wastewater and make their way to municipal wastewater treatment plants (MWWTPs). Over the last ten years, scattered reports from Europe and North America have appeared with increasing frequency demonstrating that residuals of PPCPs are being detected in wastewater and MWWTP effluents (Ternes, 1998; Golet *et al.*, 2001). Discharges from MWWTPs represent one of the largest sources of effluent to the Canadian environment (Chambers *et al.*, 1997)

Aquatic life in waters receiving MWWTP effluent is continuously exposed to low levels of PPCPs. The active ingredients in PPCPs are different from many conventional pollutants because they have been designed to elicit a biological response at low levels. This characteristic and the potential for continuous exposure of aquatic ecosystems to PPCPs have generated concern that PPCP releases may be harmful.

Most PPCP risk assessment work to date has been done in Europe. Environmental concentrations of PPCPs in North America may be different from Europe as a consequence of

differing PPCP use patterns, water consumption and MWWTP operations. The degree of reduction may depend on the design and operational characteristics of the MWWTP (e.g. temperature, solids retention time (SRT)). The physical and chemical properties of these compounds will have an impact on their capacity to be reduced within the MWWTP. Possible fates of PPCPs and their metabolites within a MWWTP are: (1) mineralization to CO_2 and water: (2) retention to the solids portion (sludge/ biosolids) if the compound entering the plant or the product of biologically mediated transformation is lipophilic; (3) release to the receiving water either as the original compound or as a degradation product.

The goal of this study was specifically to expand and in some cases establish a Canadian database for the presence of selected acidic drugs, triclosan, polycyclic musks, and selected estrogens in MWWTP influent and effluent. It also examined how treatment configuration (e.g. lagoons, conventional activated sludge (CAS), and CAS followed by media filtration (CAS + filtration)) and operational parameters may impact reduction. The acidic pharmaceuticals and musks were selected on the basis of levels of use in Canada, reported aquatic toxicity effects, and the ability to analyze for the compounds at low levels. The results of this study are expected to provide initial information for aquatic toxicologists considering the Canadian situation.

2. Materials and Methods

2.1 Description of Municipal Wastewater Treatment Plants (MWWTPs)

Twelve Ontario MWWTPs discharging into the Thames River were chosen as the sample sites. The Thames River, the second largest river in southwestern Ontario, flows into the Great Lakes System. The selection of MWWTPs along a single water course facilitated a companion study of upstream and downstream concentrations of selected pharmaceuticals and endocrine disrupting compounds (D. Bennie, personal communication).

The flows treated by the 12 selected plants ranged from 1,200 to 105,000 m^3/day and the residential wastewater contribution was 40-100% of the total flow (Table 1). The treatment types represented by the twelve plants include: lagoons with and without media filtration of the effluent (plants numbers 1, 2, 3); CAS (7 plants; 8 process streams, plants 4-10); and CAS + filtration (2 plants, plant numbers 11 and 12). Phosphorous removal was achieved by the addition of either ferrous chloride (plants 7, 9, and 11) or alum (plants 1-6, 8 and 12), with addition typically occurring at the head of the plant. Disinfection was not practiced by any of the plants during the sampling period. Treatment plant characteristics and operational parameters during the survey are presented in Tables 1 and 2. In addition, Table 2 presents selected indicators of influent and effluent quality. The degree of nitrification, as presented in Table 2, is based on effluent values as not all organic nitrogen present in the influent is available to the nitrifying organisms.

2.2 Sample collection

Samples were collected from October to December of 2002. A 24-hour composite sample was taken for three consecutive weekdays for all CAS and CAS + filtration locations, and all lagoon influents. At the MWWTPs employing lagoons (plants 1, 2, 3), effluent grab samples of 18L were taken from the holding ponds.

At the continuous discharge plants (plants 4 - 12), influent wastewater samples were collected downstream of grit removal. Influent and effluent samples were collected concurrently with no compensation for the hydraulic residence time (HRT) of the process. Samples were 24 hour equal-volume composites with 350 mL being collected every 30 minutes. Influent sampling at plant 12 occurred at a higher frequency (175 mL every 15 minutes) due to an influent stream with a highly variable character caused by intermittent industrial contributors (e.g. livestock abattoir and dairy processing plant). Effluent samples were collected as close as possible to the discharge point for all plants.

Sigma 900 refrigerated autosamplers (American Sigma, Loveland, Colorado) were used for all composite sample collection. All composite and grab samples were collected into 20 L stainless steel canisters. The stainless steel containers were cleaned with Contrad 70 detergent (Decon Laboratories Inc., Bryn Mawr, PA) and rinsed sequentially with reverse-osmosis water and methanol (Caledon Laboratories, Georgetown, Ontario) prior to use.

Samples were transported from the field to the laboratory on ice within 5 hours of the completion of sampling. Aliquots for acid pharmaceuticals, musk analysis and conventional analytes were generated at the time of sample delivery.

2.3 Sample Preparation and Chemical Analysis

In this study, the term "acidic drugs" refers to a group of pharmaceuticals that are extractable at a pH of 2 or less. Acidic analytes included: salicylic acid (SYL), ibuprofen (IBP), gemfibrozil (GMF), naproxen (NPX), ketoprofen (KTP), diclofenac (DCF), indomethacin (IND), clofibric acid (CLF), fenoprofen (FNP) and fenofibrate (FNF). Samples analyzed for acidic drugs and triclosan (TCL) were extracted and prepared for analysis as described previously (Lee *et al.*, 2004a). Briefly, samples were filtered through 1.2 μm glass fibre filter paper (Whatman GF/C) then acidified and spiked with a recovery standard. The spiked filtrate was applied to pre-conditioned reverse phase solid phase extraction (SPE) cartridges and extracted on a vacuum manifold. The SPE cartridges (Oasis HLB 6 mL, 200 mg) were purchased from Waters (Mississauga, ON, Canada). The SPE cartridges were extracted with methanol (Caledon Laboratories, Georgetown, Ontario). The extract was solvent exchanged, derivatized with N, O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS) purchased from Supelco (Oakville, ON, Canada). The derivative was analyzed using a Model 6890 gas chromatograph equipped with a 7683 automatic liquid sampler and a 5973 Mass Selective Detector (Agilent Technologies, Mississauga, ON) following the temperature profile detailed in Lee *et al.* (2004a).

The recovery standard for these analyses was 2,3-dichlorophenoxyacetic acid (2,3-D) (Sigma-Aldrich, Mississauga, ON, Canada), prepared in acetonitrile. No deuterated or C^{13} versions of the compounds of interest were available at the time of analysis. Recovery of (2,3 D) for raw wastewater samples averaged 76% with relative standard deviation (RSD) of 20%. More consistent recoveries were achieved with treated effluent samples where the average recovery was 76% with a RSD of 11%.

Presented results were not corrected for incomplete recovery. Analytical standards for the acidic drugs were purchased from Sigma-Aldrich (Oakville, ON, Canada). All solvents were products of Caledon or Burdick and Jackson. After the study was completed, a statistically-derived method detection limit (MDL) was determined (Tables 3 and 4) using the MISA protocol (MISA, 1993).

Samples were analyzed for five polycyclic musks: Celestolide (ADBI), Phantolide (AHMI), Traseolide (ATII), Galaxolide (HHCB) and Tonalide (AHTN). Samples were extracted and prepared for analysis as described previously by Lee *et al.* (2004b). Briefly, aliquots for musk analysis were filtered using 1.2 μm glass fibre filters (Whatman GF/C) and subject to liquid/liquid extraction with petroleum ether (Caledon, Georgetown, ON). Extracts were cleaned up on deactivated silica gel and analyzed using GC/MS techniques.

No recovery surrogate was used during musk analysis. Spiked distilled water samples were used to generate percent (%) recovery. In replicate analysis of four samples, the recovery of the musk

compounds was greater than 87%. After the study was completed, a statistically derived MDL was determined using the MISA protocol (MISA, 1993).

Analysis for 17β-estradiol (E2) and estrone (E1) in wastewater has been described previously (Lee and Peart, 1998). Briefly, aliquots for these parameters were filtered, applied to conditioned SPE cartridges, and eluted with acetone. Extracts were derivatized and analyzed using GC/MS. The limits of detection shown in Tables 3 and 4 were arrived at through consideration of the instrument detection limit and the levels of chemical noise (interference) in real world samples.

Samples for conventional parameters were preserved and analyzed within the time limits specified in *Standard Methods* (APHA ,1998). The conventional parameters analyzed were: 5-day carbonaceous biological oxygen demand ($cBOD_5$); chemical oxygen demand (COD); ammonia-nitrogen (NH_3-N); nitrate-nitrogen (NO_x-N); total Kjeldahl nitrogen (TKN); total suspended solids (TSS), and total phosphorus (TP). All analysis was done as specified in *Standard Methods* (APHA, 1998).

3. Results

3.1 MWWTP Performance – Organic and Nitrogen Removal

In general, the organic strength and nitrogen content of the wastewater was typical of a weak to medium strength wastewater as defined by Metcalf and Eddy Inc (1991). The influent of plants 2-11 are of weak to medium strength. Plants 1 and 12 received wastewater with a much higher

organic strength – possibly a consequence of receiving wastewater generated by milk processing. The pH of the influents to the 12 plants ranged from 6.0 to 8.2.

At the time of sampling, all of the plants except Plant 1 were producing an effluent meeting or exceeding the general quality guidelines of 25 mg/L $cBOD_5$, 25 mg/L TSS, and 1 mg/L TP. Plant 1 produced an effluent with a TSS of 31 mg/L and a $cBOD_5$ of 35 mg/L. The pH of the effluents ranged from 6.0 to 8.6. No process upsets (e.g. shock loadings, hydraulic upsets) were noted during the sampling period.

SRTs presented in Table 2 are estimates derived using influent and effluent organic strength; aeration tank residence times and mixed liquor suspended solids values; and published kinetic and stoichiometric coefficients. The following equation was used:

$$X = \frac{Y(S - S_0)}{1 + b\theta_C}\left(\frac{\theta_C}{\theta}\right)$$

where: X is the reactor mixed liquor volatile suspended solids (MLVSS) (mg/L)

Y is the yield coefficient (0.45 mg VSS/ mg COD)

b is the decay coefficient (0.05 d^{-1})

S is the effluent COD concentration (mg/L)

S_0 is the influent COD concentration (mg/L)

Θ_C is the solids retention time (SRT) (d)

Θ is the hydraulic residence time (HRT) in the aeration tanks (d)

3.2 Acidic Drugs and Triclosan in MWWTP Influents and Effluents

The concentrations of acidic drugs in the influents and effluents of the MWWTPs were determined on three consecutive days. Influent and effluent values for all PPCP analytes are reported in Tables 3 and 4. The tables show the number of samples with concentrations greater than the MDLs. Reported medians and means were derived using all values greater than the MDLs.

IBP, GMF, NPX, KTP, DCF, and IND were measurable in the majority of samples. Three compounds were not measurable: CLF, FNP and FNF. SYL was detected in all influent samples; however, the recovery and consistency of the results were highly variable.

One of the objectives of this study was to establish and expand the Canadian database on the presence of PPCPs in municipal wastewater. Using the data generated by this study and available information on populations served by the plants, the generation rates per capita were estimated for selected acidic drugs and estrogens (Table 5). This analysis could only be completed for analytes having influent concentrations higher than the MDL. Two plants have been excluded from the analysis. One plant had extremely high per capita wastewater generation rates suggesting an error in the population value. The second plant had a portion of the flow diverted due to construction. Table 5 summarizes the median and first and third quartile values for IBP, GMF, NPX, DCF, and E1. The values generated by this estimate are appropriate for use in screening level exercises. Figures 1 and 2 illustrate the data generated for IBP and E1. Both

data sets reasonably estimate a normal distribution. Examination of the data did not suggest lower or higher overall generation rates for any specific plant.

In the WWTP effluents, many acidic pharmaceuticals were at less than measurable values. Table 4 presents the median, mean and maximum of the measurable values. Influent and effluent values were used to generate reduction values (Table 6) when analyte values were available. Average values for each plant are presented in Table 6. In some instances, reductions on two days were very similar in value and the value on the third day appeared extraneous. There was no statistical justification for excluding the third value so it was included.

Table 6 also presents information on three other scenarios: non-measurable values of the analyte in both the influent and effluent; measurable values of the PPCP in the influent and non-measurable concentrations in the effluent; and non-measurable values of the analyte in the influent and measurable values of the analyte in the effluent. These three different scenarios are represented symbolically in Table 6 and are included to assist in the interpretation of the numerical data. For instance IBP and NPX have a combination of high numerical values and there are many instances of measurable levels in the influent and non-measurable in the effluent. This would suggest that actual reductions are even greater than the reported values.

Review of the reduction data shows consistently high reductions for IBP and NPX for CAS plants. Reductions for IBP and NPX by CAS plants were in the range of 94-98% and 79-98% respectively. In a number of cases, the effluent concentrations of IBP and NPX was non-measurable and thus for practical purposes the reduction was essentially 100%. Operational

parameters may affect reduction but given the consistently high treatment efficiencies, the uncertainty in analytical measurements, and fluctuations in operating parameters over the survey period, performing a regression analysis is limited in its value.

Comments on the effect of operational parameters on the reduction of KTP and IND are difficult to make due to the scarcity of the data set. In many instances, these compounds were present in the influent at less than the method detection limit (MDL). Medians of KTP and IND reduction efficiency data are 44% and 22%, respectively.

The GMF and DCF reduction data sets are more populated than those for IND and KTP. GMF and DCF have median reductions of 66% and -34% respectively. In the DCF data set there are several negative reduction values suggesting that DCF may be deconjugated under certain conditions.

Treatment plants were grouped according to SRT: 3-5 days (Group 1), 10-15 days (Group 2), and 30 days and more (Group 3). The SRT groups and reduction data for GMF and DCF are presented in Figure 3. Inspection of this figure shows that on 12 of 26 sampling occasions DCF concentrations increased from influent to effluent. Reduction does not appear to be strongly influenced by SRTs up to 15 days, while SRTs over 30 days were associated with more frequent non-measurable effluent levels of DCF. GMF reduction appears to follow a similar pattern: plants in Group 3 achieve more non-measurable effluent concentrations while plants in Groups 1 and 2 do not shows overall trends. Higher reduction rates do not appear to be correlated with the degree of nitrification (data not shown).

TCL, an antibacterial agent, was measurable in all influent samples (Table 3) and approximately 60% of the effluent samples (Table 4). Reduction rates for the twelve plants ranged between 74-98%, with a median reduction of 93% (Table 6). The three lagoon systems produced effluents with less than measurable concentrations of TCL. The effect of SRT was not evaluated due to the high reduction efficiencies.

3.3 Polycyclic Musks in MWWTP Influents and Effluents

Synthetic musks are used as fragrances and fixatives in a number of personal care products (e.g. shampoos, skin care products, soaps). Five polycyclic musks were analyzed in this survey; the influent and effluent concentrations are presented in Tables 3 and 4. HHCB and AHTN are present at much higher concentrations than the other musks.

Reduction values for four polycyclic musks are presented in Table 6. AHMI has been excluded from this table because it was not present in measurable quantities in any of the effluents. Inspection of Table 6 shows that lagoon systems were the most effective type of treatment for reduction of musks. In the lagoon systems, HHCB and AHTN had median reductions of 99% and 98% respectively, and non-measurable effluent levels of the other three musks. Reductions for CAS and CAS+filtration were lower and more variable.

To examine the effect of SRT on treatment efficiency, plants were placed in one of three categories as above: 3-5 days (Group 1); 10-15 days (Group 2); and 30 days and more (Group 3). The SRT group and reduction values for ADBI and ATII are illustrated in Figure 4, and

those for HHCB and AHTN are presented in Figure 5. Non-measurable effluent levels of analyte and high reduction efficiencies were found for the lagoon systems in Group 3. Influent concentrations of musks for these systems were comparable to those for the mechanical plants. In contrast, for the continuous flow plants in Group 3, reduction values for HHCB and AHTN were either low or negative. The data do not support increasing reduction efficiencies with increasing SRT.

3.4 Estrogens in MWWTP Influents and Effluents

The estrogens E2 and E1 were the parameters detected with the greatest variability. E2 was detected in influent samples at concentrations ranging from 0.0061 µg/L to 0.139 µg/L. E1 was measurable in all influent samples with the exception of Plant 9, with concentrations ranging from 0.016 µg/L to 0.049 µg/L. E1 was detected in a limited number of effluent samples from CAS systems and not detected in effluent samples from lagoon or CAS+filtration systems. Generation rates per capita for E1 were calculated in a similar fashion as for acidic drugs. Median values along with the upper and lower quartile values are presented in Table 5. Generation rates per capita were not calculated for E2 because of the high proportion of non-measurable values in the influents. Reduction values for plants having a measurable concentration of E1 are included in Table 6. Typical reductions are on the order of 80%. The value of 3% reported by Plant 7 seems to be extreme given the other values but there is no obvious reason for this value. Neither SRT nor degree of nitrification appear to be correlated with degree of reduction.

4. Discussion

4.1 Acidic Pharmaceuticals and Triclosan

Pharmaceuticals enter MWWTPs as either the original compound or as one of its possible metabolites. Typically, drug metabolism within the human body involves Phase I and Phase II reactions. Phase I reactions usually convert the parent drug into a more polar metabolite, and include reduction, oxidation, and hydrolysis of the parent compound. Phase II reactions, also known as conjugations, involve the addition of glucuronic acid, sulphuric acid or acetic acid to the Phase I metabolite. The purpose of these metabolic reactions is to increase the ability of this compound to be excreted from the body. Drugs do not necessarily go through both Phase I and Phase II reactions.

For many reasons, it is not possible to measure glucuronides and other Phase I and II products in a wastewater matrix at present. If Phase I and II products are transformed during the treatment process to liberate the original compound then stated reduction numbers will underestimate actual reductions. If these transformations occur during collection and transport to the WWTP, then reduction values are more reflective of treatment capabilities.

Based on the liberation of conjugated estrogens in collection and treatment systems, deconjugation of acidic drugs within these systems seems possible. D'Ascenzo *et al.* (2003) examined the fate of natural estrogens (e.g. conjugated estriol, estradiol, and estrone) from source to MWWTP influent. In the original wastewater free E2 and E1 were not detected. At the entrance of the MWWTP, free estrogens and sulfated estrogens were the dominant species suggesting that deconjugation of glucuronides occurred during storage and transport in the

collection system. Ternes *et al.* (1999b) demonstrated that glucuronide cleavage of hormones could be achieved by aerobic activated sludge cultures. It seems reasonable, given the previous observations, to expect some deconjugation of acidic drugs within the collection and treatment system of the observed 12 WWTPs used in this study. The highest degree of deconjugation would be expected to occur in extended aeration and lagoon systems due to their long contact with the bacteria within the system. Reductions are based on measured influent and effluent concentrations of the analyte in the liquid phase. Processes that may influence the apparent reduction include: sorption and de-sorption from the primary sludge and biosolids, complete and partial biodegradation, and generation through deconjugation of human metabolites.

There are now three Canadian studies characterizing acidic pharmaceuticals in MWWTP influents and effluents: Metcalf *et al.* (2003), Miao *et al.* (2002) and this study. Overlapping analyte lists allows comparisons between studies. Metcalf *et al.* (2003) characterized plants across Canada during the fall (October-December 2000). The results presented by Metcalf (2003) are comparable with those presented here in the influent results for FNP, GMF, KTP, and DCF; however, results for NPX reported by Metcalf *et al.* (2003) are consistently higher. For example, in this study NPX concentrations in the influent range between 2.3 µg/L and 17.1 µg/L, while Metcalf *et al.* (2003) report a value of 40.7 ug/L.

In an earlier survey, three Ontario MWWTPs were surveyed by Miao *et al.* (2002). The limits of detection in effluents were between 0.005 and 0.010 µg/L for most of the selected acidic pharmaceuticals. Measured values for DCF, FNP, GMF, IND, and NPX were in the range of

0.025µg/L to 0.050 µg/L. CLF and KTP were not detected. In our survey CLF was consistently not measurable and KTP was not measurable in 31 of 36 samples.

The acidic pharmaceuticals considered in this survey were often effectively reduced by the MWWTP. Those detected most consistently: IBP, NPX, GMF, and DCF, can be grouped in pairs based on their patterns of reduction. The first pair, IBP and NPX, had consistently high reductions (86-99%). Any effects of treatment configuration and operational characteristics were not easily seen. Ternes *et al.* (1999b) has shown that that approximately 15% of IBP is excreted unchanged or as its glucuronide metabolite; the remaining percentage is allocated to other metabolites such as hydroxy-ibuprofen, carboxy-ibuprofen and their respective conjugates. The high reduction cited here refers only to IBP and excludes any metabolites. In the literature, reduction values for NPX as reported by Ternes (1998) and Stumpf *et al.* (1999) are 66% and 78%, respectively. NPX has been noted to be biodegradable (Khan and Ongerth, 2002) and has a sufficiently high K_{ow} to make sorption to solids a possible reduction mechanism.

The second pair, GMF and DCF, had generally lower reductions and more inter-plant variability. Reduction of GMF as reported in this survey ranges from -127% to 71% with a median reduction of 55%. In the literature, reported reduction values for treatment plants are 46% (Stumpf *et al.*, 1999) and 69% (Ternes, 1998). Eight of the plants in this survey showed DCF reductions in the -143% to 77% range. Ternes (1998) reported a reduction of 69% while values of 53 to 74% have been reported by Clara et al. (2003) for a number of different sampling campaigns. Zwiener *et al.* (2001) reported reductions of 1 to 6% for a pilot MWWTP. The low reductions reported by this investigator may be a consequence of a selected culture with minimal ability to

degrade DCF due to culture conditions. Poiger *et al.* (2000) compared MWWTP influent and effluent concentrations, with no compensation for the plant hydraulic residence time, and found DCF reductions of 5 to 50%. Clara et al (2003) noted DCF removals of 7-29% in laboratory experiments using activated sludge systems fed a synthetic wastwater spiked with pharmaceuticals. Campanella *et al.* (2003) presented an average reduction of 30% and commented on the high degree of scatter present in their reduction data set (<10 to 88%).

The data set obtained from this survey also contained a high degree of scatter. In addition to the somewhat variable composition of municipal wastewater from day to day, the variability of the analytical results could be in part due to matrix effects in the samples. For example, extraction recoveries were lower for raw influent, and co-elution from the SPE cartridges could have caused the derivitization efficiency to drop. The post-derivitization step, which includes a back extraction with petroleum ether and dichloromethane, caused a heavy emulsion with most raw influents, creating the potential for further losses of analytes. For subsequent work the emulsion problem was solved through the use of sodium sulfate.

TCL is an antibacterial compound widely used in personal care products. This compound is of particular interest due to the large quantities consumed; its potential to inhibit bacterial activity in the receiving water; and the formation of toxic compounds such as 2,8-dichlorodibenzo-p-dioxin and 2,4-dichlorophenol, in addition to oligomerization and dissolved organic matter-coupled products during photolysis (Latch *et al.*, 2003). Canadian data on treatment plant releases are limited to this study and that of Lee et al (2004a). Influent and effluent data derived from these two studies are comparable. Effluent values from two Louisiana treatment plants

range from 0.010 -0.021 µg/L (Boyd et al., 2003); these values are consistent with the reported Canadian values of 0.03 to 0.074 µg/L. MWWTP effluents with up to 0.65 µg/L of TCL have been noted for European plants (Paxeus, 1996; Lindstrom *et al.*, 2002).

Singer *et al.* (2002) provide influent and effluent TCL levels for seven different Swiss plants: five conventional plants achieving organic carbon removal and nitrification and two biological nitrogen removal plants. In addition to biological treatment, effluents were subject to flocculation and filtration which would result in further reductions of TCL if it was particulate bound. Effluent values of TCL ranged between 0.042 µg/L and 213 µg/L based on the analysis of samples collected in a flow proportional manner. Differences in the sampling regime and use patterns could be responsible for differences between Swiss and North American studies.

Reductions for the twelve plants in this study range from 90 to 99% based on analysis of soluble TCL in influent and effluent samples (Table 6). Singer *et al.* (2002) and Bester (2003) provide the only other published removal data for TCL and both report reductions exceeding 90%. Singer *et al.* (2002), based on data from a one week survey, suggest that the fate of TCL is: 79% biologically degradation (i.e. mineralization or transformation); 15% sorption to wasted sludge; and 6% discharged to the receiving water.

4.2 Musks

Polycyclic musk compounds are the fragrance components of many personal care and cleaning products and their release to the aquatic environment is primarily through MWWTP discharges. Musks are of interest because they have been shown to bioaccumulate in fish (Herberer *et al.*, 2001; Metcalf *et al.*, 2003). Heberer (2002) suggests that with knowledge derived from risk

assessment studies, risks to humans and fish-eating predators can almost be excluded; however, more information and research is needed concerning the formation, identification and possible toxicological and endocrinal effects of metabolites from polycyclic musk compounds.

Effluent values from this survey can be compared to two other Canadian investigations. Slightly lower values are reported by Ricking *et al.* (2003); this could be due to dilution since the samples were collected from the effluent pipe at a water depth of 1 m. Lee *et al.* (2004b) characterized secondary clarifier effluent (Table 7) and the median values of that survey are very close to those of this study.

Heberer (2002) reviewed effluent polycyclic musk concentrations originating from a number of German plants, finding that the German data were higher, but still comparable, with those in a UK STP data (Simonich *et al.*, 2002). Noser et al (2000) reported levels of HHCB and AHTN of 2.98 µg/L and 2.04 µg/L, respectively, in Swiss MWWTPs. A comparison of the Canadian database with European studies indicates that in general polycyclic musk concentrations in Canadian MWWTP effluents are 5 to10 times lower. More extensive European and Canadian databases would be useful in confirming this initial observation.

4.3 Estrogens

Servos *et al.* (2005) examined selected Canadian MWWTPs and found average influent values for E1 and E2 of 0.049 and 0.016 µg/L, respectively, which were somewhat higher than the average values obtained from this study (E1 0.0295 µg/L and E2 0.0083 µg/L). Effluent values presented by Servos *et al.* (2005) for E1 are also 50% higher than those reported in this study.

After sampling effluents of ten Ontario WWTP in the late fall (November), Ternes *et al.* (1999a) found the median values of E1 and E2 to be 0.003 µg/L and 0.006 µg/L respectively. This is consistent with the median value for E1 of 0.008 µg/L as determined by this study.

In the investigation carried out by Servos *et al.* (2005) reductions in E2 ranged from 40% to 99% and E1 reductions from -46% to 95%. In our study, all plants showed essentially complete removal of E2, since all effluent results showed non-measurable effluent levels of E2. Reductions of E1 were on the order of 80%.

5.0 Summary

The goals of this study were: (1) to expand and in some cases establish the Canadian database of influent and municipal wastewater treatment plant effluent concentrations for selected PPCPs; (2) to determine if three treatment configurations (e.g. lagoons, conventional activated sludge (CAS), and CAS followed by media filtration (CAS + filtration)) affect PPCP reduction; (3) explore whether CAS operational parameters could be correlated with reduction. The following conclusions can be drawn from this survey:

- IBP and NPX had consistently high reductions. Median reductions for IBP and NPX were 95% and 93%, respectively. No direct correlation was found with any particular operational parameter.
- Comments on the reduction of KTP and IND are difficult to make due to the scarcity of the data set. In many instances, these compounds were present in the influent at less than the method detection limit (MDL). Medians of KTP and IND reduction efficiency data were 44% and 30%, respectively.

- GMF and DCF have median reductions of 66% and -34%, respectively. Several negative reduction values in the data set suggest that DCF may be deconjugated under certain conditions. A graph of DCF reduction showing reductions for all days indicates DCF generation in twelve of twenty six sampling occasions. Reduction does not appear to be strongly influenced by SRT.
- Triclosan had a median reduction efficiency of 93%. No direct correlation was found with any particular operational parameter.
- In general, lagoon treatment systems are the preferred treatment configuration for reduction of acidic drugs and triclosan, generating effluents with less than measurable levels of these compounds. Influent values of these compounds were comparable to those received by other treatment configurations.
- A comparison between Canadian values and those of European studies indicate that in general polycyclic musk concentrations in Canadian MWWTP effluents are 5-10 times lower. More extensive European and Canadian databases would be useful in confirming this initial observation.
- Median reductions for ADBI and ATII were 39% and 43%, respectively. The reduction efficiencies of ATII varied widely over the three day sampling period for a single plant. Within-plant consistency increases with SRT above the SRT range of 3-5 days. Higher reductions of ATII appear to be co-incidental with longer SRTs for the 3-5 day category.
- The same within-plant trend seen with ATII is present in the graphs of HHCB and AHTN reductions. Median reductions for these two compounds are approximately 40%. Lagoon systems (which have long SRTs) have essentially complete reduction while continuous

flow systems do not. A better understanding of musk reduction mechanisms could provide insight.

- In general, lagoon treatment systems are the preferred treatment configuration for reduction of musk concentrations in municipal wastewater.
- E2 appears to be essentially removed from all continuous flow activated sludge treatment systems and lagoons; E1 reductions were also high (80%).

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Table 1. Characteristics of 12 municipal wastewater treatment plants along the Thames River, Canada.

Plant Number	Treatment Mode	Primary Clarification	Average daily flow (m^3/day)*	Wastewater Sources	Significant Industrial Discharges	Media Filtration
1	Lagoon		1,458	70% Res 30% IC	Feed mills, cheese factory	No
2	Lagoon		2,256	85% Res 15% IC	None	Yes
3	Lagoon		1,215	80% Res 20% IC	None	Yes
4	Activated Sludge	Yes	5,506	100% Res	None	No
5	Activated Sludge	Yes	19,260	90% Res 10% IC	University, 2 hospitals	No
6	Activated Sludge	Yes	105,300	55% Res 45% IC	None	No
7	Activated Sludge	Yes	15,300	80% Res 20% IC	None	No
8a	Activated Sludge	Yes	1,984	40% Res 60% IC	Recycled paper processor, meat packers, automotive, landfill leachate	No
8b	Activated Sludge	Yes	3,967		None	No
9	Activated Sludge	Yes	17,994	40% Res 60% IC	Metal working	No
10	Activated Sludge	Yes	3,585	35% Res 65% IC	Auto parts, animal feed	No
11	Activated Sludge with Filtration	Yes	20,716	50% Res 50% IC	None	Yes
12	Activated Sludge with Filtration	No	3,592	55% Res 45% IC	Animal feed, dairy, abattoir	Yes

* Average flows during the sampling period
Res: residential
IC: industrial / commercial

Table 2. Operational parameters and characteristics of influent and effluent streams for 12 municipal wastewater treatment plants along the Thames River, Canada.

Plant Number	Solids Retention Time (days)	Mixed Liquor Suspended Solids (mg/L)	Plant Hydraulic Retention Time (hours)	Influent 5-day Biochemical Oxygen Demand (mg/L)	Influent Total Suspended Solids (mg/L)	% Nitrified effluent[+]	Influent Temperature (°C)	Effluent Temperature (°C)
1	N/A			532	330	N/A	6	4
2	N/A			147	64	N/A	3	6
3	N/A			129	116	N/A	-2	2
4	5.5	2836	15	205	155	89	16	16
5	6	2084	22	128	108	90	18	17
6	5	2450	16	127	199	75	11	4
7	5.5	1743	15	154	108	71	6	4
8a	4.5	2482	15	113	227	90	19	18
8b	3	3030	10	113	227	64	19	17
9	10.5	2105	13	51	108	66	15	17
10	30	4554	23	178	137	84	7	1
11	15	3280	22	71	78	94	11	14
12	N/A	5171	43	409	191	89	19	11

[+] % Nitrified effluent = (effluent nitrate *100)/(effluent Total Kjeldahl Nitrogen + effluent nitrate)
N/A = not applicable

Table 3. Influent concentrations (µg/L) of acidic drugs, triclosan, polycyclic musks and hormones for 12 municipal wastewater treatment plants along the Thames River, Canada. (N=36)

Analyte (µg/L)	Method Detection Limit	n < MDL	Median	Mean	Maximum
Salicylic Acid	0.087	0	14.1	13.7	27.8
Ibuprofen	0.061	0	8.84	8.45	16.5
Gemfibrozil	0.077	2	0.418	0.453	0.965
Naproxen	0.074	0	5.22	5.58	17.1
Ketoprofen	0.088	23	0.136	0.146	0.289
Diclofenac	0.062	0	0.140	0.204	1.01
Indomethacin	0.10	21	0.196	0.230	0.64
Fenofibrate	0.026	36	-	-	-
Fenoprofen	0.066	36	-	-	-
Clofibric Acid	0.066	36	-	-	-
Triclosan	0.031	0	1.86	1.93	4.01
Celestolide	0.016	10	0.0345	0.0372	0.067
Phantolide	0.018	29	0.0220	0.0420	0.150
Traseolide	0.013	7	0.131	0.168	0.723
Galaxolide	0.012	0	1.701	2.031	5.20
Tonalide	0.0085	0	0.687	0.804	2.00
Estradiol *	0.005	11	0.0081	0.0083	0.0139
Estrone *	0.005	0	0.0302	0.0295	0.0487

Note for Estradiol and Estrone 31 and 33 samples were analysed, respectively.

Table 4. Effluent concentrations (µg/L) of acidic drugs, triclosan, polycyclic musks and hormones for 12 municipal wastewater treatment plants along the Thames River, Canada. (N=39)

Analyte (µg/L)	MDL	n<MDL	Median	Mean	Maximum
Salicylic Acid	0.087	36	0.104	0.106	0.121
Ibuprofen	0.061	23	0.353	0.384	0.773
Gemfibrozil	0.077	22	0.255	0.246	0.436
Naproxen	0.074	18	0.351	0.452	1.189
Ketoprofen	0.088	30	0.114	0.125	0.210
Diclofenac	0.062	15	0.140	0.194	0.748
Indomethacin	0.10	30	0.149	0.190	0.507
Fenofibrate	0.026	39	-	-	-
Fenoprofen	0.066	39	-	-	-
Clofibric Acid	0.066	39	-	-	-
Triclosan	0.031	15	0.106	0.108	0.324
Celestolide	0.016	28	0.0200	0.0207	0.09
Phantolide	0.018	39	-	-	-
Traseolide	0.013	13	0.047	0.0453	0.072
Galaxolide	0.012	0	0.876	0.0751	1.82
Tonalide	0.0085	1	0.298	0.274	0.600
Estradiol *	0.005	34	-	-	-
Estrone *	0.005	17	0.013	0.0076	0.038

* Note for Estradiol and Estrone 34 and 34 samples were analysed, respectively.

Table 5. Estimated per capita generation rates of acidic pharmaceuticals (mg/capita·day) based on data from 12 municipal wastewater treatment plants along the Thames River, Canada.

	Ibuprofen	Gemfibrozil	Naproxen	Diclofenac	Estrone
Mean	2.50	0.20	2.27	0.088	12.8
Relative Std Deviation	44%	68%	36%	82.9%	35%
Median	2.51	0.189	2.31	0.075	11.8
1st Quartile	1.86	0.091	1.94	0.047	9.95
3rd Quartile	3.46	0.251	2.72	0.093	16.5

Table 6. Percent reduction (%) of acidic drugs, triclosan, musks and estrogens by 13 municipal wastewater treatment trains, Thames River, Canada.

Plant	Treatment Type	IBP	NPX	GMF	KTP	DCF	IND	TCL	ADBI	AHMI	ATII	HHCB	AHTN	E1	E2
1	Lagoon	###	###	-127	*##	###	*##	###	***	**#	**#	95	87	###	###
2		###	###	**#	***	###	###	###	###	**#	###	99	98	86(##)	###
3		###	###	###	***	###	***	###	###	*##	###	99	99(#)	###	###
Median												99	98	86	###
4	CAS	95	79	43(#)	***	-88	15(#*)	74	66	###	81	67	70	###	###
5		98(##)	96(#)	###	***	30	**+	98	36(#)	***	73	67	63	83(##)	###
6		###	95	69	***	22	**+	97	###	***	9	54	67	82	###
7		94	86	38	9+	-103	-41	93	33(#)	***	64	46	51	3	###
8a		98	98	###	44	###	**#	93	###	***	69	39	37	++	###
8b		94	81	66	-133	28	30(#,*)	85	41	***	56	15	30	+++	###
9		91	90	71	**#	-143	**#	89	11(##)	***	25	20	13	50	###
10		###	98(#)	###	###	77	63(+#)	98	***	***	*	9	-1	80(##)	###
Median		95	93	66	44	-34	23	93	39		65	43	37	80	###
11	CAS+filtration	###	###	###	**#	9	**#	95	*##	***	56	31	44	###	###
12		###	###	###	***	###	*##	###	***	***	***	-7	-67	###	###
Median								95			56	19	12		

(*) – indicates the number of times non-measurable values present in the influent and effluent
(+) – indicates the number of times non-measurable values present in the influent and measurable values present in the effluent
(#) – measurable in the influent and non-measurable in the effluent
N/A – analysis not completed on influent
Abbreviations: IBP = Ibuprofen, NPX = Naproxen, GMF = Gemfibrozil, KTP = Ketoprofen, DCF = Diclofenac, IND = Indomethacin, TCL = Triclosan, ADBI = Celestolide, AHMI = Phantolide, ATII = Traseolide, HHCB = Galaxolide, AHTN = Tonalide, E1 = Estrone, E2 = Estradiol

Table 7. Polycyclic musk concentrations (after Lee *et al.*, 2004b)

Analyte (µg/L)	Median	Range
Celestolide	0.016	0.014-045
Phantolide	0.005	0.005-0.013
Traseolide	0.054	0.008-0.203
Galaxolide	0.915	0.825-1.570
Tonalide	0.422	0.337-0.661

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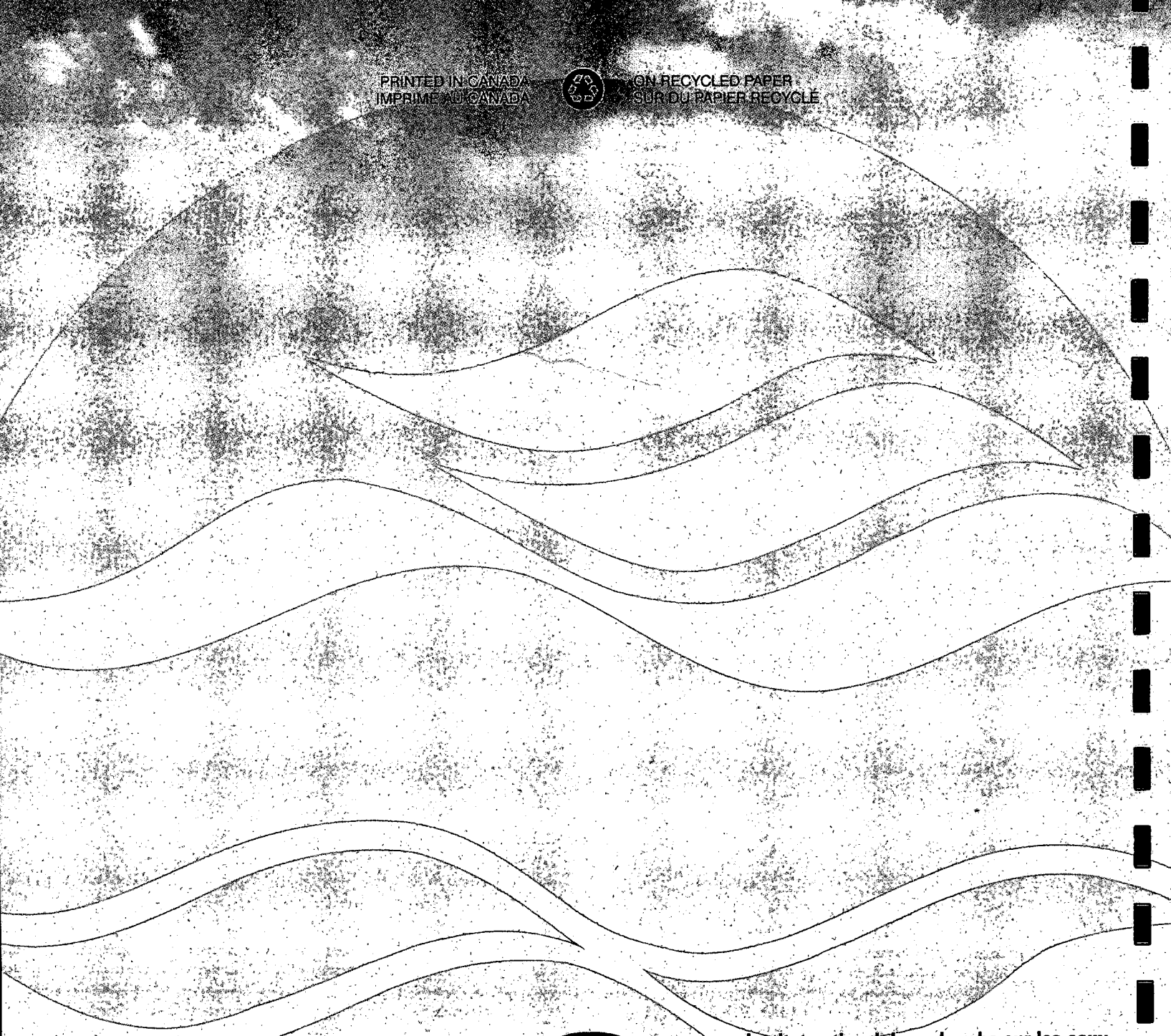

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