Occurrence and removal of illicit drugs in different wastewater treatment plants with different treatment techniques

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Research

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Abstract

Background: As a class of contaminants of emerging concern (CECs), illicit drugs should be taken into account in the water management because of their social and public health risks. Wastewater treatment plants (WWTPs) are usually considered as the source and sink of contaminants, and drug residues are observed in their effluents due to the inefficient removal of CECs. In this study, wastewater samples were collected from eight WWTPs in Changzhou City for assessment of the abuse of 12 illicit drugs in the city by wastewater-based epidemiology (WBE) method. Results: Drug concentrations ranged from

Background

The consumption of illicit drugs has been a growing concern worldwide. World Drug Report of 2018 points out that more than 275 million residents around the world were taking drugs, with thousands of tons of illicit drugs consumed annually [1]. In China more than 2.4 million residents consumed drugs at least once in 2017 [2]. Ultimately, illicit drugs enter the wastewater system as unaltered forms or/and metabolites. These drugs were excreted after human consumption, or they were released illegally after police intervention [3]. Studies on the occurrence of target compounds in raw waters from wastewater treatment plants (WWTPs) supply invaluable information on illicit drug using and consumer trends at local, national and international levels [3-6]. The method called “wastewater-based epidemiology (WBE)” [4, 7-10] could assess the consumption of illicit drugs by local people, which has been confirmed by traditional approaches [11]. Limited researches have been conducted to investigate the occurrence of illicit drugs in the influents and effluents in WWTPs in China [5,7,8].

Illicit drugs may cause potential risk to aquatic organism and human health when they enter the natural water environment [7,8,12]. The effluents from WWTPs were the principal source of illicit drugs in the surface water [13]. The elimination efficiency of illicit drugs varied compound by compound in WWTPs. For instance, the removal rate of amphetamine (AMP) and cocaine (COC) was more than 90%, while the removal efficiency of 3,4methylenedioxymethamphetamine (MDMA) or methamphetamine (METH) was below 50% [14-17]. The majority of illicit drugs cannot be completely removed in the traditional WWTPs, and a great number of illicit drugs were able to get in the surface water, groundwater and drinking water sources with the discharged effluent [18].

In the present study we reported the detection of 12 illicit drugs in both influents and effluents from eight WWTPs using different treatment techniques in Changzhou city, east China. To our best knowledge, this is the first detailed investigation on the occurrence and removal of illicit drugs in different WWTPs with different treatment techniques. The total consumption amounts of target drugs in the sampling period were estimated using WBE. Furthermore, the removal rates of target analytes in the WWTPs were assessed, and the effect of various wastewater treatment techniques on the drugs removal was determined as well.

Materials And Methods

Reagents and chemicals

The target illicit drugs, including methamphetamine (METH), cocaine (COC), 3,4-methylenedioxymethamphetamine (MDMA), benzoylecgonine (BE), amphetamine (AMP), methadone (MET), norketamine (NK), codeine (COD) , ketamine (KET), 3,4-methylenedioxyamphetamine (MDA), methcathinone (MC), heroin (HR), and methamphetamine-d_8 (METH-d_8) with purities greater than 98.2%, were purchased from Cerilliant Corporation (Round Rock, TX) (Fig. 1). Formic acid (FA) (99%) was obtained from Sigma-Aldrich (Steinheim, Germany) and ammonium hydroxide (NH_3H_2O) was obtained from J&K Scientific (Beijing, China). HPLC-grade methanol (MeOH) and acetonitrile (ACN) were obtained from Fisher Scientific (Poole, UK). Ultrapure water was acquired using a Milli-Q ultrapure water system (Millipore, Bedford, MA, USA).

Sample collection

Wastewater samples (all samples were collected on the same day and at 60 minute intervals (24 h composites) in a time scale) were taken from 500 mL both influents and effluents of 8 WWTPs in Changzhou. The main information of each WWTP was shown in Table 1. All samples were stored in polyethylene terephthalate bottles (Adjust all water samples to pH=2, then add 5 ng METH-d8 and EDTA-2Na) and transferred to laboratory in 4^oC ice box, and treated within 24 h.

Sample preparation and extraction

The solid phase extraction (SPE) method was used to extract illicit drugs in the samples according to our previous work [12,19]. Briefly, wastewater sample was filtered through glass microfiber filters (GF/C Whatman, 0.47μm), followed by SPE with Oasis MCX cartridges (60 mg, 3 cc) through a vacuum extraction manifold. With the addition of internal standards, wastewater samples were loaded on the cartridges which were preconditioned with successive 5 mL of methanol and 5 mL ultrapure water. After water loading, the MCX cartridge was rinsed by 6 mL ultrapure water, and drained by a vacuum for half an hour for removing excessive water. The target drugs were eluted from the cartridges by 6 mL 5% NH_3H_2O in ACN. The extract was concentrated with weak N_2 stream in the water bath at 40^oC to dryness, and the volume was brought to 1 mL with 10% acetonitrile in water. Prior to the analysis with ultra-performance liquid chromatography equipped with tandem mass spectrometry (UPLC-MS/MS), the extracts were passed through a 0.22 μm nylon syringe filter.

Instrumental analysis

Quantification of analytes was conducted with a Waters triple quadrupole mass spectrometer equipped with an electrospray ionization source under positive electrospray ionization (ESI+) and multiple-reaction monitoring (MRM) mode. The analytes were analyzed within one week after sample extraction and were separated with an Acquity UPLC BEH C18 column (2.1 mm × 50 mm, 1.7 μm particle size), and the column temperature was kept at 40^oC during sample analysis. The Milli-Q water with 0.1% FA was used as mobile phase A, and the ACN as mobile phase B. The velocity of flow was maintained at 0.45 mL/min, and the volume of injection was 5 μL. The elution gradient was starting with 98% of A for 0.5 min, then increased A to 50% gradually within 4.5 min, and then increased A to 98% at 4.6 min. The next process was to maintain A at 98% for 1.4 min, and then decreased A to 2% gradually within 6.2 min, and kept constant for 1.3 min to re-equilibrate before the injection of the next water sample. The total working time was 7.5 min. The ion spray

voltage was 4.5 kV. The source temperature was 450°C. The collision gas pressure was 9.0 Psi. Other experimental parameters were listed in Table S1.The software MassLynx and TargetLynx V4.1 was used to acquire and process the data.

Quantification and quality control

To determine the accuracy and precision of the analytical approach, Milli-Q water and wastewater samples spiked with deuterated standards were treated with the same pretreatment method. Three types of water (tap water, Milli-Q water and surface water) were spiked with 10 illicit drugs and 2 metabolites at 10 ng/L to evaluate the recoveries from different matrices. Each sample was analyzed in triplicate to calculate the relative standard deviation (RSD), and non-spiked sample was analyzed to measure the background concentrations simultaneously. The relative recoveries ranged from 73.8% to 92.6% in tap water, 71.2% to 85.1% in Milli-Q water, and 71.8% to 87.4% in surface water (Table S2). For spiking at different concentrations (0.1, 10, and 50 ng/L) in wastewater, the recoveries ranged from 70.7% to 95.1%. RSDs≤10% were obtained for each compound (Table S3). Matrix effect was investigated by spiking the deuterated standards (5μg/L) into wastewater samples before extraction. A 8-point calibration standard curve (0.1-10 μg/L) was conducted with correlation coefficient (R^2) greater than 0.99. A procedural blank and a quality control sample (2μg/L) were injected between every 10analyses. In the procedural blank the concentrations of target analytes were below the detection limits. The limit of detection (LOD) was calculated by the minimal detectable content (at signal/noise ratio of 3) of a drug from the 12 environmentally matrix-spiked extracts. The LODs for each compound were calculated by MassLynxand TargetLynx V4.1 (Waters, US) from the relative recoveries data of each compound, which were ranging from 0.005-0.025 ng/L, with details featured in Table 2.

Wastewater based epidemiology

Wastewater based epidemiology (WBE) was an effective method to investigate the chemical consumption of the urban population. Combined with traditional survey method, WBE can provide real-time and objective data for drug related policy. WBE assesses daily per capita consumption of drugs by the measured concentration of target residues in population and wastewater, daily flow rate of WWTPs, and human metabolic factors (corrected relationship between target residues and actual drugs) in the service area of WWTPs [7].

The daily mass load of each compound into a certain WWTP was calculated with the equation as follows: (see Equation 1 in the Supplemental Files)

The influent concentration of the illicit drug was experimentally determined. Average influent flow on every sampling day was provided by each WWTP. Similarly, WWTPs supplied population from serving community.

Results And Discussion

Illicit drugs in wastewater

Concentrations of 12 illicit drugs in both influents and effluents from 8 WWTPs were summarized in Table 3. METH, COC and KET were the most frequently observed compounds, which were found in all influent samples. The detection frequencies of AMP, MC, COD, HR, MET, MDMA, and MDA in influents were greater

than 75%, whereas BE was only detected in the WWTP TNQ-2 (Table 3). METH concentrations ranged from 1.28 to 51.62 ng/L and from <LOD to 22.4 ng/L in the influents and effluents, respectively. The lowest and highest average concentrations of METH were observed at WJQ-1 (1.20 ± 0.5 ng/L) and XBQ-1 (51.62 ± 3.5 ng/L), respectively. In general, METH concentrations in the present study were similar to the influent levels from WWTPs at Nanjing, Shanghai and Nanning cities in China [8], slightly higher than in eastern Canada and Dutch sewage water [20], slightly lower than in UK WWTPs [21], and much lower than in WWTPs in Yinchuan, Xiamen and Shenzhen in China [7].

AMP concentrations were lower than LOD at XBQ-1, and the highest AMP concentration was detected at JTQ-1 (10.29 ± 1.2 ng/L). Positive correlations were discovered between METH and AMP concentrations (r=0.88). AMP is the main metabolite of METH and also the ingredient of Selegiline, the medicine for treating Parkinson's disease [9]. Studies have shown that the conversion ratio of METH to AMP after metabolism is between 4% and 7% [7]. It is logical to draw that when the concentration rate of AMP to METH was between 0.04-0.1, AMP was mostly derived from the transformation of METH. The ratio of concentration of AMP to METH was greater than 0.1 at most WWTPs in this study, indicating that the emergence of AMP was more likely to be related to the use of prescription drug Selegiline. This contrasted sharply with the status in European countries where more AMP relative to METH use was detected [22-24].

KET was observed in eight influent samples, and its concentrations were ranging from 0.78 - 2.50 ng/L, while NK was not detected in the influents (Table 3 and Fig. 2). Although KET was widely present in all WWTPs, the degradation product NK could not be detected. Compared with other cities in China, NK was consistently observed in WWTPs in the southern cities of Shenzhen and Guangzhou, with concentrations higher than in Beijing and Shanghai [10], implying the different consumption patterns of KET in different areas of China.

MDMA was observed in 75% of the samples. And the concentrations of MDMA ranged from 0.18-0.55 ng/L in influents and 0.18-0.67 ng/L in effluents. These concentrations were lower than many reported levels in countries and regions with high consumption of MDMA, such as the Taiwan (≤LOD-4.82 ng/L) [25], US (70 ng/L) [26], UK (13.9 ng/L) [19] and Spain (180 ng/L) [27]. Compared with those cities, Changzhou is a relatively small area with less entertainment places, and the detected MDMA concentrations were nearly 100 times lower than in European countries. Similar to MDMA, low concentration of MDA was observed for 0.24-4.85 ng/L. The concentration range of COC is 0.18-0.75 ng/L, which is far lower than the influent concentration of UK WWTPs (5.1-208.9 ng/L) [28] and Canada WWTPs (289-823 ng/L) [29]. A comparative study was conducted in 9 WWTPs in Beijing, Guangzhou, Shenzhen and Shanghai in China [30], indicating that the illicit drug use pattern in China was different from European countries. In China, the use of METH and KET was the main concern, while the most popular ones in European countries were COC and MDMA.

MET was observed in more than 50% of both influents and effluents wastewater samples. Concentrations of MET in the influent were in the range of ≤LOD-1.24 ng/L and in the effluent for ≤LOD-0.18 ng/L. In this study, the concentrations of MET were lower than the reported levels in other countries, such as the US (62 ng/L) [9], Croatia (94 ng/L) [12] and Belgium (16 ng/L) [10].

Removal of illicit drugs from WWTPs

The removal efficiency of target analytes depends strongly on the wastewater treatment techniques. A summary of the removal efficiency of each WWTP was presented in Fig. 3. In this study, the treatment techniques of the eight WWTPs included anaerobic anoxic oxic (A^2/O) process, anoxic oxic (A/O) process and sequencing batch reactor activated sludge (SBR) process. The main advantages of the SBR process were fast sedimentation rate, high reaction rate, and good degradation performance for refractory organics. The SBR was therefore more effective for removing illicit drugs in the treatment process, while the A^2/O and A/O processes were mainly used to remove organic N and P. SBR process was able to effectively remove illicit drugs from water, with the highest removal rate for the total 12 target drugs of 79%, followed by the A^2/O process (73%). In the SBR process, illicit drugs were firstly eliminated in the primary sedimentation tank, which was likely related to the treatment conditions such as ambient/water temperature, sewage water composition or biological population [9]. The activity of the biomass in the SBR process accelerated the decomposing of the organic pollutants, resulting in the removal efficiency up to more than 70%.

Most of the investigated illicit drugs and their metabolites are hydrophilic, therefore they are expected to be predominantly present in the dissolved aqueous phase and to adsorb poorly onto solid particles. However AMP and METH were highly resistant to the biological degradation and they were mainly removed from wastewater by sorption [31-33]. The negative removal of some illicit drugs was also observed in the WWTPs, with the highest negative removal rates of MC and MDMA up to -155.9% and -60.7%, respectively. Negative removal was also reported for KET [20, 24, 32] and other target drugs [9], which may be bound up with the increased transformation of precursor compounds or parent compounds, hydraulic residence time, and/or desorption from suspended solids in the wastewater treatment processes [23, 35]. Since the influent and effluent samples were not collected on the base of the hydraulic residence time of the WWTPs, the higher concentration found in the effluent for some drugs might be due to the fact that the corresponding influent load was different.

BE was detected only in the influent of TNQ-2. BE is the major metabolite of COC. COC concentrations were relatively low in all 8 wastewater treatment plants, and the removal rate of COC in the sewage treatment process was basically below 50% [27]. This result was inconsistent with the previous study that COC was easily metabolized into BE, and after human body metabolism, only 1% will be excreted from urine in the form of COC, and 25%-45% was excreted in the form of BE [4, 36]. METH, MET and MDA were effectively removed in different WWTPs, with removal rates ranging from 51.3% to 100%. Conversely, MDMA displayed negative removal in four WWTPs in this study. The composition and physicochemical properties was shown in Table S4. This phenomenon was possibly related to its highly recalcitrant property in wastewater and longer half-life of MDMA compared with other target drugs, which may elevate both the persistence of MDMA in wastewater and resistance to biological processes in the secondary treatment procedures [37].

Mean loads of illicit drugs

Among the 8 WWTPs, LYS-1 receives domestic sewage, XBQ-3 and JTQ-1 receive both domestic sewage and industrial sewage, and the rest 5 WWTPs receive industrial sewage only.

The mass loads of illicit drugs were ranged from 0.01 mg/d/1000 inh (MDMA in LYS-1) to 20.65 mg/d/1000 inh (METH in XBQ-1) (Fig. 4). Average loads of METH, AMP, KET, COD, MDA and MC in the eight WWTPs were

5.89±8.74, 0.84±0.62, 0.34±0.41, 0.48±0.76, 0.42±0.39 and 0.41±0.36 mg/d/1000 inh, respectively (Table 3).

High METH loads were found in XBQ-1 (20.65 mg/d/1000 inh) and XBQ-3 (18.99 mg/d/1000 inh). These loads were lower than those of wastewater samples (1500-1800 mg/d/1000 inh) in Australia [6] and other cities in China such as Haerbin (181.20±6.50mg/d/1000 inh) [8], Yinchuan (148.00±145.20mg/d/1000 inh) [8, 12], and the mean METH load in WWTPs from18 cities in China was 67.80±45.2 mg/d/1000 inh, which was 3-fold higher than the mean load in this study [8].

COC and MDMA consumption was much lower than METH in this study, ranging from 0.02 to 0.15 mg/d/1000 inh for COC and 0.02-0.16 mg/d/1000 inh for MDMA. The consumption of COC and MDMA estimated in the present study was lower than 20.00-200.00 mg/d/1000 inh and 5.00-50.00 mg/d/1000 inh, respectively, in WWTPs of South-East Queensland in Australia. The average loading of MC was < LOD -0.92 mg/d/1000 inh, which was lower than 0.50 mg/d/1000 inh in Great Britain and 1.0 mg/d/1000 inh in Italy [36]. The average loading of MET was 0.09±0.06 mg/d/1000 inh, far lower than Sweden (0.50-29.00 mg/d/1000 inh) [38] and Finland (1.2-9.5 mg/d/1000 inh) [24].

The average daily load of MDA and MDMA was < LOD - 1.10 and < LOD - 0.16 mg/d/1000 inh, respectively. The measured level was comparable with other Chinese cities for Shenzhen and Guangzhou [10], but was much lower than the MDMA load of 5-41 mg/d/1000 inh in the working days of the 25 WWTPs in France [6]. The maximum load of HR was 0.98 mg/d/1000 inh, which was much lower than the average HR load of 61 mg/d/1000 inh in the Ebro River Basin [9]. HR load in Guangzhou and Shenzhen in China was less than 4.0 mg/d/1000 inh [12], while the HR load in western cities of China was higher. The daily load of COD was < LOD - 2.01 mg/d/1000 inh, which was basically consistent with the average COD load of 5.7 mg/d/1000 inh in South Korean [28], far low than the UK [27], with corresponding load of 565 mg/d/1000 inh.

Although COD and MET were detected in most WWTPs in this study, the detection concentrations were significantly lower than European cities with the estimated COD load of 2-1998 mg/d/1000 inh [3]. MET and COD are used as controlled medicines in China [39] (In 2018, the State Drug Administration listed COD as the prohibited substance for adolescents and children). In Europe however, MET is used as an alternative medicine for methadone oral solutions. In most EU countries, COD can be used legally. Drugs containing codeine are approved by national procedures and sold as prescription or over-the-counter drugs in different prescriptions [39].

Conclusions

In the present study, we investigated the occurrence and elimination of 12 illicit drugs and their metabolites from 8 wastewater treatment plants in Changzhou City. METH, AMP, KET, MDMA and MET were the dominant drugs in these WWTPs, with higher concentrations than other drugs. Among the treatment techniques, SBR process was efficient for removal of illicit drugs, while the removal efficiency of target drugs by A/O process was low. Results from wastewater-based epidemiology showed that METH was the most consumed illicit drug, which should draw much attention by regulatory and management communities.

Abbreviations

METH: methamphetamine, COC: cocaine, BE: benzoylecgonine, MDMA: 3,4-methylenedioxymethamphetamine, AMP: amphetamine, MET: methadone, NK: norketamine, COD: codeine, KET: ketamine, MDA: 3,4-methylenedioxyamphetamine, MC: methcathinone, HR: heroin, WWTPs: wastewater treatment plants, A^2/O: anaerobic anoxic oxic process, A/O: anoxic oxic process, WBE: wastewater based epidemiology

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and material

The datasets used and analyzed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

YD, CG and HZ involved in the experiments and manuscript writing, XY, LC and DW were responsible for the data analysis. JX contributed to the study design and manuscript correction. All authors read and approved the final manuscript.

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Tables

Table1 The main information of 8 WWTPs

	TNQ-1	TNQ-2	XBQ-1	XBQ-2	XBQ-3	WJQ-1	JTQ-1	LYS-1
Treatment process	SBR process	SBR process	A^2O	A\O	A^2O	A\O	A^2O	A^2O
Average water intake (m^3/d)	15000	25000	160000	3000	80000	25000	35000	70000
People (million)	0.08	0.30	0.30	0.01	0.15	0.13	0.25	0.65
Influent characteristics								
COD (mg/L)	1123	780	255	1185	260	971	265	247
N (mg/L)	21.46	18.74	36.1	88	31.54	12.3	35	39.62
P (mg/L)	4.65	2.08	4.41	-	4.46	3.02	3.42	6.95
NH_4 (mg/L)	21.46	18.74	23	88	27.3	12.3	30	28
Effluent characteristics								
COD (mg/L)	178	150	41	51.3	32.8	60	33	41
N (mg/L)	5.43	4.17	8.78	24.7	8.26	2	10.89	8.83
P (mg/L)	1.69	0.95	0.15	-	0.11	0.49	0.25	0.17
NH_4 (mg/L)	5.43	4.17	2	24.7	2	2	2	2

Table 2 Validation parameters - retention time (Rt), relative retention time (Rel. Rt), linearity range, correlation coefficient obtained from calibration curve, instrumental limits of detection ($LOD_{S/N}$) and instrumental limits of quantification ($LOQ_{S/N}$).

analytes	R_t (min)	Rel. R_t (min)	Linearity range (µg/L)	Calibration curve	R^2	$LOD_{S/N}$ (µg/L)	$LOQ_{S/N}$ (µg/L)
AMP	1.86	0.08	0.1-10	Y=19828.7x-554.71	0.9995	0.01	0.015
METH	2.04	0.08	0.1-10	Y=40079.6x-265.875	0.9987	0.005	0.01
MC	1.67	0.08	0.1-10	Y=36872.3x-1050.63	0.997	0.01	0.02
MDA	1.93	0.07	0.1-10	Y=11955.7x-417.15	0.9992	0.02	0.025
MDMA	2.04	0.08	0.1-10	Y=47352.2x-1614.27	0.9985	0.005	0.01
NK	2.22	0.07	0.1-10	Y=36217.3x-663.94	0.999	0.01	0.02
KET	2.29	0.07	0.1-10	Y=44710.4x+3013.45	0.9997	0.02	0.05
BE	2.27	0.07	0.1-10	Y=23465.0x-597.23	0.9988	0.005	0.01
COD	1.76	0.07	0.5-10	Y=2682.0x-131.27	0.9978	0.01	0.05
COC	2.74	0.07	0.1-10	Y=21003.5x-10.74	0.997	0.025	0.02
MET	3.99	0.08	0.1-10	Y=27369.3x-4660.57	0.9988	0.01	0.02
HR	2.64	0.07	0.1-10	Y=803.6x-55.84	0.995	0.02	0.05

Table 3 Concentration and frequency of detection of the target illicit drugs and their metabolites in the influent and effluent (ng/L).

Compound		TNQ-1	TNQ-2	XBQ-1	XBQ-2	XBQ-3	WJQ-1	JTQ-1	LYS-1	Freq. (%)
METH	influent	5.13	1.95	51.62	1.21	35.61	1.20	6.58	45.08	100.00
	effluent	0.38	0.47	16.59	□LOD	8.24	0.74	19.71	22.44	87.50
AMP	influent	8.11	1.43	□LOD	2.22	2.82	1.19	10.29	4.27	87.50
	effluent	□LOD	1.03	1.06	□LOD	□LOD	1.06	1.22	1.29	62.50
KET	influent	1.72	0.78	0.78	0.90	2.50	1.05	0.87	0.96	100.00
	effluent	□LOD	0.19	0.86	0.35	1.21	0.70	0.94	1.01	87.50
NK	influent	0.38	□LOD	0.73	0.34	□LOD	0.23	0.16	□LOD	62.50
	effluent	0.58	0.27	0.36	0.31	□LOD	0.18	0.33	0.18	87.50
COD	influent	10.71	□LOD	□LOD	0.80	0.81	0.40	0.42	0.94	75.00
	effluent	0.00	0.42	0.34	0.61	□LOD	0.46	0.30	0.39	75.00
HR	influent	0.00	0.38	0.30	0.74	0.37	0.46	6.99	□LOD	75.00
	effluent	1.53	0.34	0.42	0.69	0.34	0.43	1.81	0.40	100.00
MET	influent	0.44	1.24	0.14	0.60	□LOD	0.72	□LOD	0.09	75.00
	effluent	□LOD	0.09	□LOD	0.19	□LOD	0.10	□LOD	0.10	50.00
COC	influent	0.75	0.38	0.18	0.51	0.18	0.30	0.19	0.21	100.00
	effluent	0.49	0.19	0.18	0.40	0.18	0.20	0.19	0.19	0.00
BE	influent	□LOD	0.19	□LOD	□LOD	□LOD	□LOD	□LOD	□LOD	12.50
	effluent	□LOD	□LOD	□LOD	□LOD	□LOD	□LOD	□LOD	□LOD	□LOD
MDMA	influent	0.48	0.00	0.30	0.55	0.00	0.21	0.26	0.18	75.00
	effluent	0.67	0.18	0.49	□LOD	0.21	0.22	□LOD	0.19	75.00
MDA	influent	1.75	0.24	2.74	□LOD	□LOD	1.10	4.85	2.18	75.00
	effluent	0.60	□LOD	0.24	□LOD	0.46	0.25	2.36	0.39	75.00
MC	influent	0.61	0.28	2.16	0.95	□LOD	2.69	6.58	1.62	87.50
	effluent	0.50	0.67	0.18	1.23	0.31	0.19	□LOD	0.58	87.50

Figures

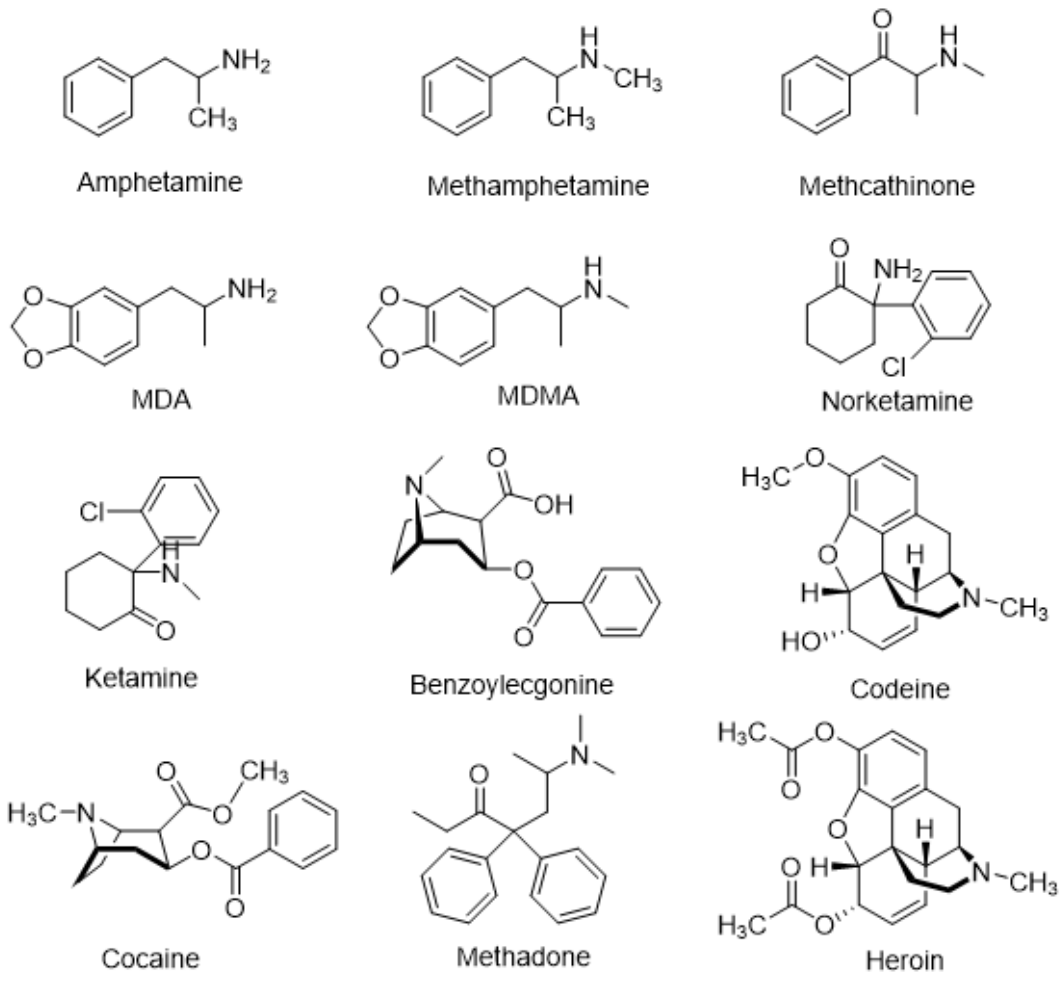

Figure 1

Structure of 12 target illicit drugs and their metabolites in this study

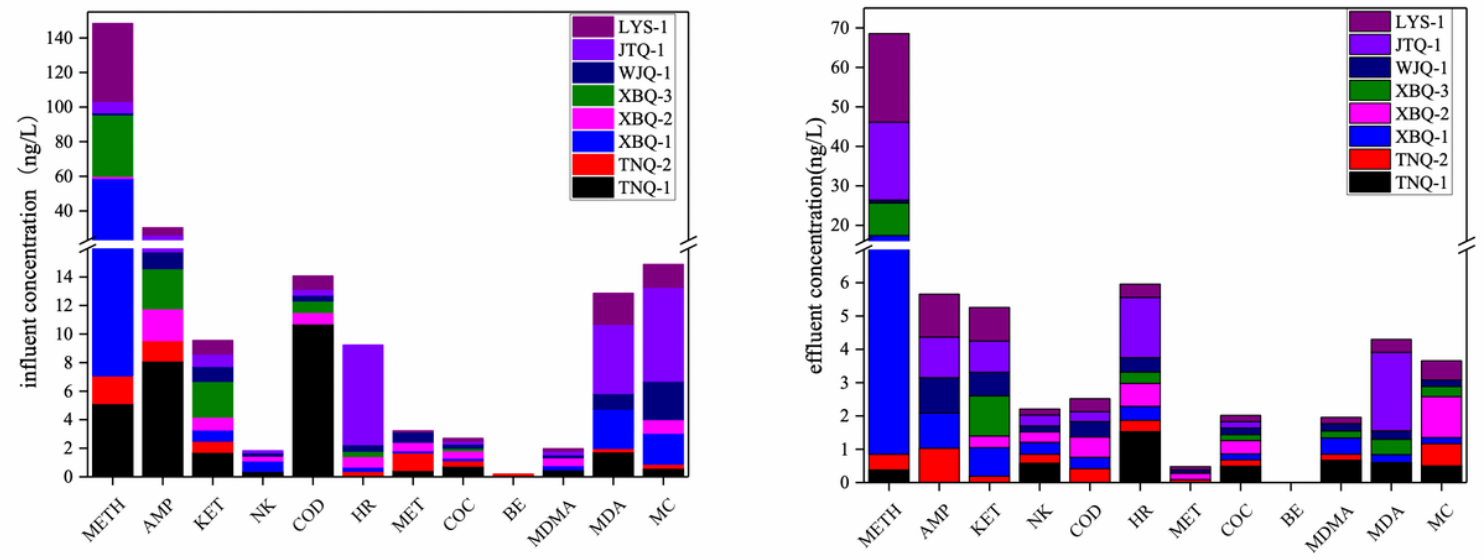

Figure 2

Average concentrations of target analytes over 24 hours period in wastewater influent and effluent of 8 WWTPs.

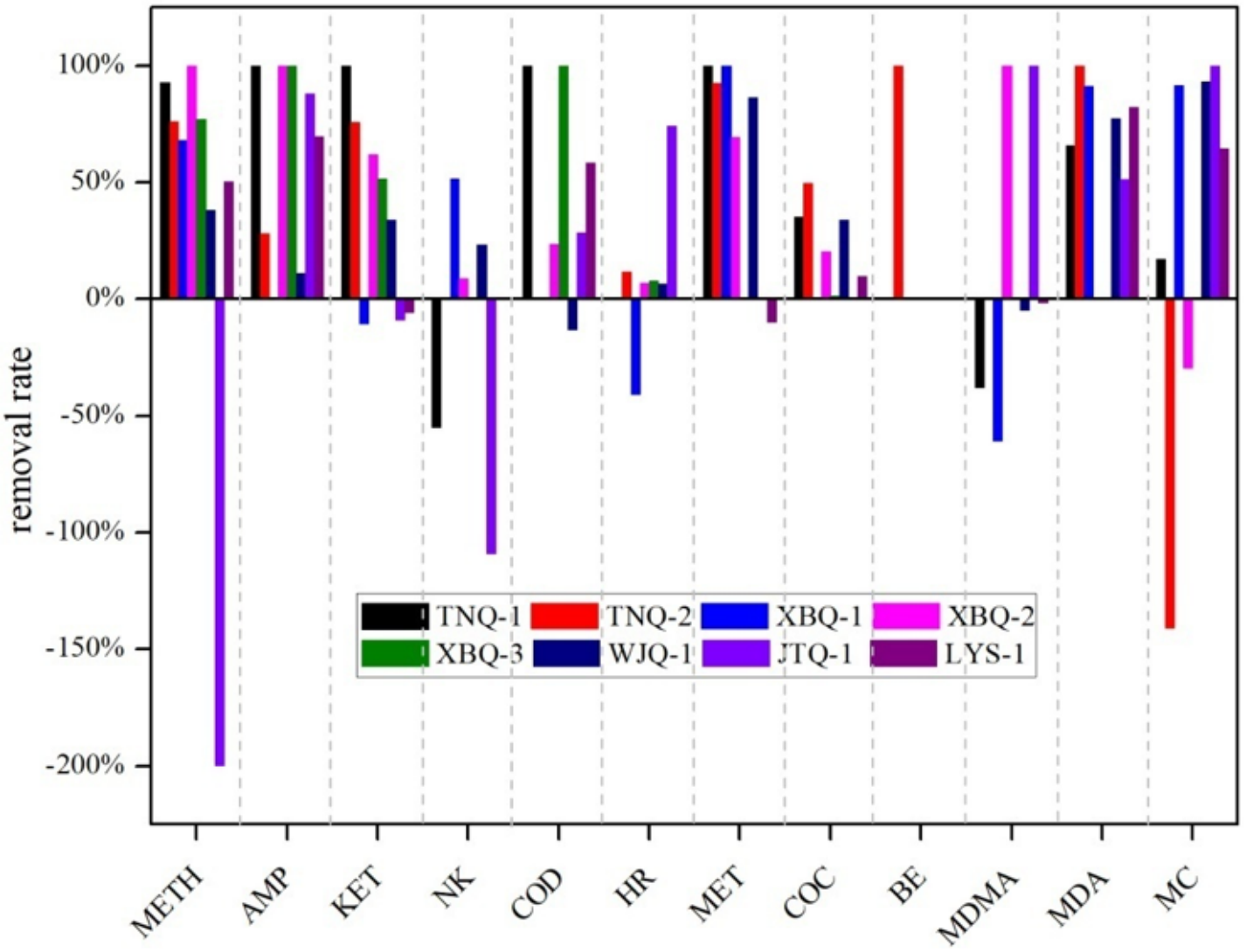

Figure 3

Comparison of removal efficiency of illicit drugs in the 8 WWTPs.

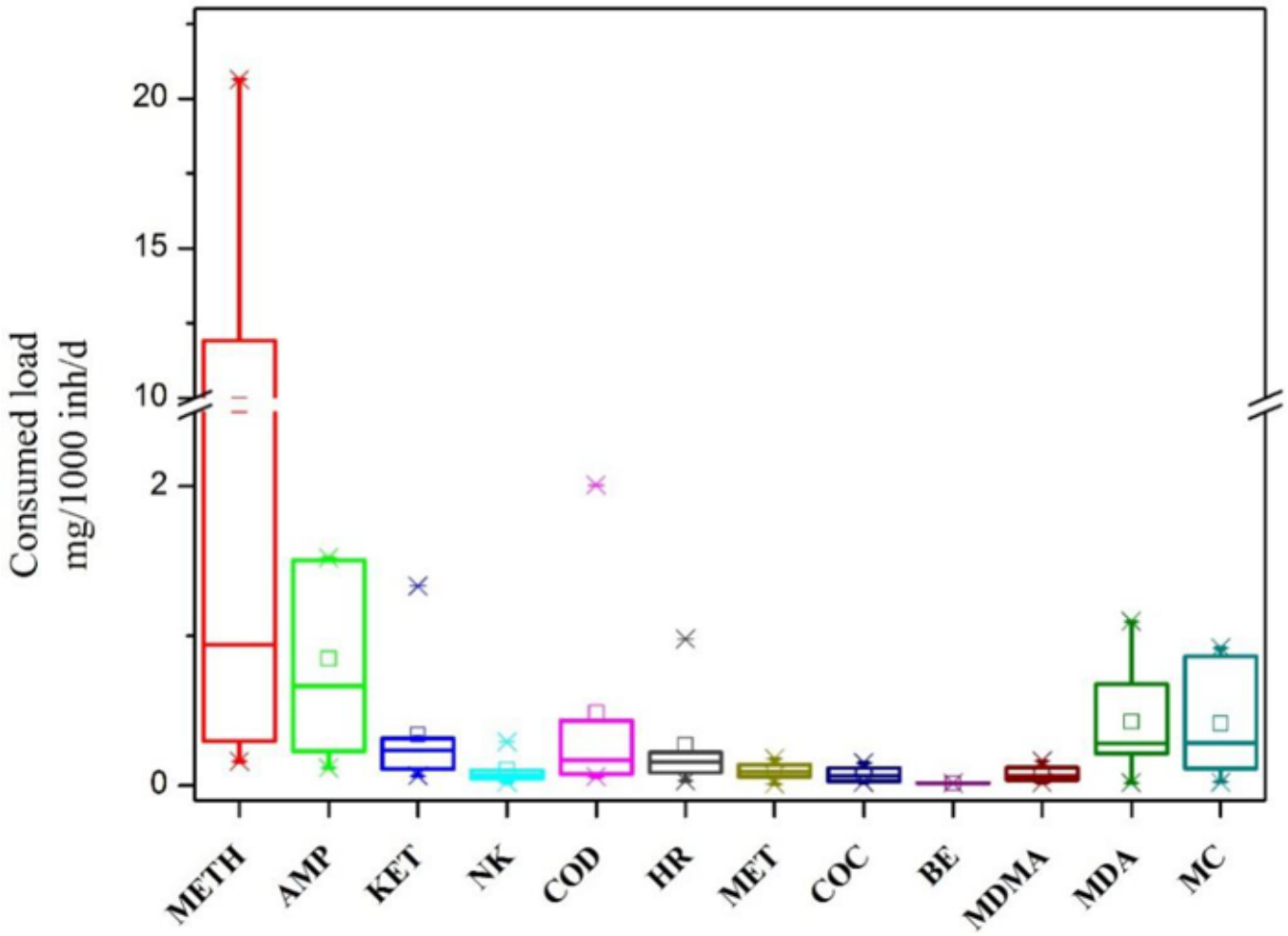

Figure 4

The estimated illicit drugs consumption per inhabitant in eight wastewater treatment plants in Changzhou City.

Supplementary Files

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- Supplementarymaterials.docx