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Discharge inventory of pharmaceuticals and personal care products in Beijing, China

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ABSTRACT

Pharmaceuticals and personal care products (PPCPs) are emerging environmental contaminants, whose potential risk for the ecological environment has caused wide attention in recent years. In China, quite a large amount of PPCPs were annually emitted into the environment. Their existence in different matrix has been reported frequently, including river water, sediment and soil. However, the contribution from different sources was seldom reported and still unclear in China. Wastewater treatment plant (WWTP) was usually considered to be the main source to the urban river, but livestock and aquaculture farms were also reported as significant pollution sources of PPCPs due to poor environmental management in China. This study summarized environmental discharges of different PPCPs from various sources and obtained the discharge data through different environment media in Beijing, the statistical source of PPCPs was analyzed in detail. The sources comprised WWTPs, excess sludge, hospital wastewater, municipal untreated wastewater, aquaculture wastewater and landfill leachate. This article helps understand the general situation and the potential risk of PPCPs in Beijing.

1. Introduction

Pharmaceuticals and personal care products (PPCPs) are used to treat or prevent human and animal disease and improve the quality of daily life [1,2]. Human-use PPCPs are mainly emitted into WWTPs and finally enter the surface water, as the effluent is used for irrigation and sludge is applied in agriculture as fertilizer [3,4]. Veterinary pharmaceuticals tend to be released to aquaculture directly and after usage in livestock, it may indirectly pollute groundwater during the land application of manure and slurry in livestock [5]. PPCPs may also be released to the aquatic environment from manufacturing sites.

China produces and consumes a large amount of PPCPs every year and a substantial amount of work has been done by the scientists as it has been reported frequently in different media with significant concentration and potential effect on human and environment [2,6–9]. They may also cause persistent contamination due to continuous discharge [4,10].

As emerging contaminants, PPCPs has currently become one of hotspots in environmental research [11–14]. However, the contribution, transport and fate of PPCPs in different environmental media and sources is unclear, which leads to an absence of comprehensive information to grasp the overall pollution situation [8,15–17]. Previous studies indicated that WWTP was the dominant PPCPs source to aquatic environment [18–21]. But other sources may contribute more in some cases [22]. Usually, the effluents from wastewater enhance the levels of pollutants, but it was shown in a research that the surface water concentration was significantly diluted by WWTPs effluent [23], the high concentration suggested there may be other point or non-point PPCPs sources existed.

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As the Chinese political and culture communication center, Beijing has developed rapidly in recent decades. With the increase of population and urbanization under speedy economic growth, many public problems such as 'City disease' as well as environmental pollution in megacity was prevalent [24]. The municipal utilities and public service facilities constructed previously could not meet the cities' requirement yet. The drainage pipeline system was mainly built in eighties and ninties of the 20th century, which does not cover the municipal network completely [25,26]. Currently, the water treatment capacity is 1.37 billion tons, far less than the total wastewater volume discharged in Beijing, which implies quite a proportion of untreated wastewater. It was reported, seven discharge outlets along the Liangshui River discharged 60,000 tons of untreated wastewater per day into the river (http://green.sohu.com/20130822/n384773793.shtml). The pollution was frequently reported, as high concentration in surface water and sewage was significantly related with livestock where PPCPs was polluted seriously (from ng/L to ug/L) [9,18,27]. Caffeine concentration in Beijing surface water was up to 4200 ng/L and the popular bug repellent DEET (N,N-Diethyl-3-methyl benzoyl amide) was 546 ng/L in summer [28,29].

For the issue above, we identified key PPCPs sources and surveyed their pollution status and contribution through literature research and statistical analysis. The sources include influent, effluent and sludge in WWTPs, untreated municipal wastewater, hospital wastewater, aquaculture wastewater, livestock excretion (dung and urine) and leachate in municipal landfill. The article established a comprehensive PPCPs' discharge inventory from multiple pollution sources and provided a scientific basis for environmental management of such PPCPs in Beijing.

2. Materials and methods

The investigation method of PPCPs contamination is shown in Fig. 1. The research launched the investigation through official statistics from official website database and statistics in literature. Original statistics mainly comprise PPCPs concentration data in different sources, parameter statistics include discharge coefficient and emission quantity from all the sources.

Recently, researchers mainly focused on the PPCPs contamination related to wastewater in WWTPs. PPCPs discharge was classified into two categories: liquid sewage poured into aquatic environment including resident sewage, factory wastewater, hospital wastewater and aquaculture wastewater; and solid waste dumped on soil, and then leached into groundwater or washed into surface water via surface water runoff, comprising sludge from WWTPs, excretion from livestock and waste solid from landfill. We assumed the influent concentration of PPCPs in WWTPs as the concentration in urban raw untreated wastewater [30,31]. In the investigation, we adopted the PPCPs concentration in influent and effluent, excess sludge, hospital wastewater and livestock wastewater as the local pollution data in Beijing, while concentration in aquaculture wastewater, landfill leachate and livestock excretion was estimated by the average concentration data at national level due to lack of local data. Meanwhile, we summed up the sample numbers of inflow and outflow and PPCPs concentration in hospital wastewater in the supporting information. Here, the PPCPs discharge from drug industries was not included due to the poor related data reported. The PPCPs pollution load in untreated wastewater was obtained via concentration of PPCPs in inflow to WWTPs. The volume of wastewater was acquired from the Beijing Statistical Yearbook[31]. In the estimate process, if the concentration data was given in scope in literature, we calculated the statistics with the mean in the estimation, we calculated using ' 1/2 LOQ (limit of quantitation)', or ' 1/2 LOD (limit of detection)' if the concentration was 'ND'. The statistics was listed in Tables S1,S3,S4. The calculation method was shown in Table S2.

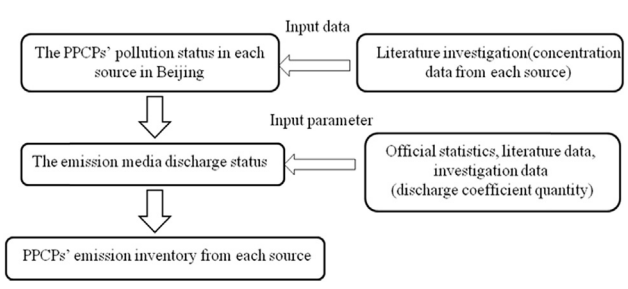

Fig. 1. Investigation method of discharge loads of PPCPs in Beijing.

On the whole, we have searched articles involving 119 PPCPs substances. The dominant group was antibiotics, followed by psychiatric drugs, anti-inflammatory drugs and personal care products. Some official statistics was calculated from China Statistics Yearbook 2013 [32], China Animal Husbandry Yearbook 2013 [33], The Chinese Fishery Yearbook 2013 [34], Chinese Medicine Statistical Yearbook 2011 [7] etc. Other statistics were sought through literature retrieval tools including Web of Science, Elsevier, Springer and Google, China National Knowledge Infrastructure (CNKI) and Wan Fang DATA etc.

Gap of discharge data related to all the pollution media was the main source of the uncertainty of the research. Besides, the detection method for different substances in different media as well as the difference of statistics method provided in the internet or reports also lead to the uncertainty. In the data processing, we had try our best to obtain a comprehensive information and reduce the miss distance between the statistics status and the actual status.

3. Results and discussion

3.1. The sources of PPCPs

The statistics results are involving 119 PPCPs pertaining to 11 categories (Table S1) in various pollution sources. The reported data related to hospital wastewater mainly focused on psychiatric drugs. Its emission quantity was classified into municipal wastewater, along with factory sewage [35]. For livestock and aquaculture farms, the statistics were mainly antibiotics [5,36,37]. Statistics concerning WWTPs comprised all kind of PPCPs categories [38–41].

3.1.1. Municipal wastewater

Generally, municipal wastewater treatment facilities collected the water via municipal pipeline from residential area, hospitals and manufacturers, when public utilities were complete [17]. In WWTPs, the municipal wastewater was processed mainly in some methods such as activated sludge method connected with oxidation ditch, biological membrane technology. In Beijing, more than 70% of sewage was treated by activated sludge method [42]. Traditional sewage treatment process including activated sludge method, oxidation ditch (OD) and biological membrane (BMR) method showed high efficiency in removing macromolecular particles in sewage, such as inorganic particles and macromolecular organic matters [1,28,43]. However, removal efficiency of PPCPs was low due to their special physicochemical characteristics of PPCPs (molecular conformation, Kow (octanol-water partition coefficient)) and WWTPs' operation condition (BOD, HRT, SRT, pH and temperature etc.) [2,44]. Actually, adsorption and biodegradation