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Optimizing sampling strategy for Chinese National Sewage Sludge Survey (CNSSS) based on urban agglomeration, wastewater treatment process, and treatment capacity

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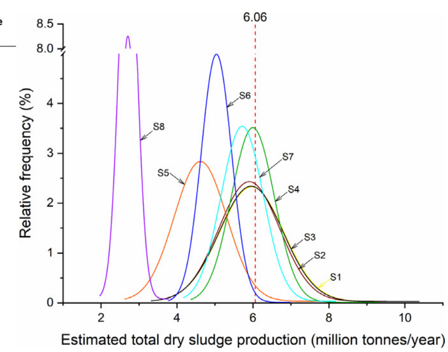
HIGHLIGHTS

- Population, GDP, and sewage treating indicators in 19 city clusters were analyzed.
- The Yangtze River Economic Belt contributes nearly 1/3 of Chinese indicators.
- Statistical sampling was developed for Chinese national sewage sludge survey.
- Stratified random sampling based on capacity provides the optimal representation.
- Excessive stratification may increase in bias.

GRAPHICAL ABSTRACT

Sampling Strategy	Stratification	Sample Size
Strategy 1	1 stratum	83
Strategy 2	20 strata by urban agglomeration	82
Strategy 3	8 strata by treatment process	82
Strategy 4	**3 strata by treatment capacity**	**82**
Strategy 5	160 strata by urban agglomeration and treatment process	70
Strategy 6	60 strata by urban agglomeration and treatment capacity	79
Strategy 7	24 strata by treatment process and treatment capacity	83
Strategy 8	480 strata by urban agglomeration, treatment process, and treatment capacity	53



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ABSTRACT

As a sink and source of contaminants, sewage sludge is a good matrix to capture the spatial-temporal trend of chemicals and assess the potential risks these chemicals pose to human health and the environment. In order to understand these chemical risks, a robust statistical sewage sludge sampling strategy for Chinese wastewater treatment plants (WWTPs) must be designed. The purpose of this paper is to develop such a sampling strategy for Chinese WWTPs which may be used optimally. Before creating the sampling design, the distribution of WWTPs was categorically analyzed. These categories include urban agglomeration, wastewater treatment process, and wastewater treatment capacity. Particular attention was given to the studying of population distribution, gross domestic product, WWTP number, wastewater treatment flow, and dry sludge production in each urban agglomeration. In addition, correlation analysis was conducted among these five indexes. Due to the heterogeneity of WWTPs, stratified sampling had to be used to homogenize the sampling units. The eight strategies proposed herein were based on simple random sampling and stratified random sampling methods. Moreover, the aforementioned three categories (urban agglomeration, treatment process, and treatment capacity) were intended to be stratification indicators. Furthermore, *Monte Carlo* simulations revealed that the treatment capacity based stratified random sampling strategy (Strategy 4) results in the optimal sample representation, with the smallest root mean square error compared to seven other sampling strategies with different strata. This optimal stratified

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sampling strategy, if employed during the Chinese national sewage sludge survey, has the potential to greatly contribute to data quality and assurance.

1. Introduction

A broad spectrum of chemicals currently being produced and consumed are often unintentionally washed down the drain and enter into municipal wastewater treatment plants (WWTPs) (Olofsson et al., 2012). As a byproduct which is produced during wastewater treatment, sewage sludge may contain a variety of chemicals when discharged (Meng et al., 2016; Olofsson et al., 2012). When sludge is disposed illegally and/or applied as soil fertilizer, these chemicals will be released into the environment as contaminants, posing risks to both the environment and human health (Chen et al., 2013; Yang et al., 2014b). On the other hand, sludge has been used as a matrix to explore the fluxes, temporal trends, and sources of chemicals discharged by human society (Feng et al., 2018; Nascimento et al., 2018). It is therefore submitted that the development of a regional or nationwide database of chemicals existent in sludge is a matter of urgency, that would assist engineers, researchers, and policymakers in developing the disposal technology necessary for effectively addressing and understanding the issue of sludge, its occurrence, fate, as well as the level of risk in respect of contaminants being present in sludge-applied soil (Meng et al., 2016; Venkatesan et al., 2015; Yang et al., 2015).

Four nationwide sludge surveys in the United States were conducted in 1982, 1988–1989, 2001, and 2006–2007, respectively, by the United States Environmental Protection Agency (U.S. EPA), which were aimed at identifying and quantifying priority contaminants in sludge (United States Environmental Protection Agency, 2009). In 2011, the European Commission's Joint Research Centre screened 114 analytes in 61 sludge samples collected from 15 European countries (Joint Research Centre of the European Commission, 2012). Similar investigations have been conducted in the United Kingdom (Stevens et al., 2003; Zennegg et al., 2013), Spain (Campo et al., 2014; Gomez-Canela et al., 2012), Germany (Gomez-Canela et al., 2012; Knoth et al., 2007), Sweden (Olofsson et al., 2012), Switzerland (Sun et al., 2011), and Australia (Clarke et al., 2008; Langdon et al., 2011). In a review from China, studies have shown 35 classes of organic chemicals consisting of 749 individual compounds and one mixture to be present in Chinese sludge since 1987, some of which examined a national survey (Meng et al., 2016). Sun et al. (2019) monitored the occurrence and composition profiles of polycyclic aromatic hydrocarbons (PAHs) in Chinese sewage sludge. Cheng et al. (2014) and Yang et al. (2014a) investigated heavy metals in sewage sludge from 107 and 58 WWTPs covering almost all provinces in China and identified their temporal or spatial trends.

The abovementioned surveys provide a plethora of information proving the existence of chemicals in sludge. However, the majority of these papers (except the fourth survey in the United States) only give cursory information on the number and locations of WWTPs sampled, and their sampling work is arbitrary and possess a lack of statistical rigor, which may result in the concentrations and compositions of contaminants investigated being biased. For example, Yang et al. (2014a) sampled sewage sludge from 107 WWTPs in 48 cities covering 31 provinces, autonomous regions, and municipalities, as well as Hong Kong, Macao, and Taiwan, without essential sampling details illustrating sample representativeness or sampling error being present. There can be no analysis without sampling being conducted (Murray, 1997). Modern sampling survey which originated in 1895 by Anders Kiaer was developed for appropriate sampling. It proved that random sampling could benefit inference from the sample to the population (Whitmore and Chen, 2013). Such random sampling includes simple random sampling, stratified sampling, systematic sampling, cluster sampling, and multistage sampling (Cochran, 1977). Among them, stratified sampling could ensure that the allocation of the sample is similar to that of the population, which can effectively improve the precision of the estimates (Jin et al., 2015). Moreover, stratified sampling was successfully employed in the fourth sewage sludge survey in the United States (United States Environmental Protection Agency, 2006). 分层抽样在美国的成功应用!

The present study aims to design an optimal sampling strategy for developing a Chinese National Sewage Sludge Survey (CNSSS). The main content of this study are to (1) examine the distribution of WWTPs by urban agglomeration, wastewater treatment process, and wastewater treatment capacity (specifically, the population, gross domestic product (GDP), WWTP number, wastewater treatment flow, and dry sludge (DS) production in each urban agglomeration were studied and discussed); (2) propose eight sampling strategies based on the principle of simple random sampling or stratified random sampling; and (3) choose and discuss on the optimal strategy by comparing their accuracy via *Monte Carlo* simulation.

2. Method

2.1. Data collection

Firstly, the list of investigated WWTPs was developed based on the 2016 *National Key Monitoring Enterprises of Wastewater Treatment Plants* (General Office of Ministry of Environmental Protection of the People's Republic of China, 2016). Secondly, all characteristic properties of the WWTPs, including the name, location, wastewater treatment process, and treatment flow (F) via the websites of national, provincial, municipal governments, and the WWTPs, as well as other online resources were collected. Thirdly, the DS production (D) was calculated by multiplying F by the DS production coefficient (k) and by 365 (days/year) for each WWTP. The k, as shown in Table S1 (S indicates the Supplementary material here and thereafter) was derived from the 2018 *Urban Drainage Statistical Yearbook* (China Urban Water Supply and Drainage Association, 2019). Finally, the populations and GDPs in each urban agglomeration as well as China were obtained from the 2018 *China Statistical Yearbook* (National Bureau of Statistics, 2019) and the *Statistical Book* of 31 provinces, autonomous regions, and municipalities.

2.2. Data classification

According to the 2016 *National Key Monitoring Enterprises of Wastewater Treatment Plants* (General Office of Ministry of Environmental Protection of the People's Republic of China, 2016), a total of 3809 WWTPs have been established in China. In addition, the Chinese central government launched 19 urban agglomerations (listed in Table S2) for economic development (Fang et al., 2016). These include the Yangtze Delta Urban Agglomeration, the Pearl River Delta Urban Agglomeration, the Beijing-Tianjin-Hebei Urban Agglomeration, the Middle Yangtze River Urban Agglomeration, the Chengdu-Chongqing Urban Agglomeration, the Middle and South Liaoning Urban Agglomeration, the Shandong Peninsula Urban Agglomeration, the West of Taiwan Strait Urban Agglomeration, the Harbin-Changchun Urban Agglomeration, the Central Plain Urban Agglomeration, the Central Shaanxi Plain Urban Agglomeration, the Beibu Gulf Urban Agglomeration, the Northern Tianshan Mountain Urban Agglomeration, the Central Shanxi Urban Agglomeration, the Hohhot-Baotou-Erdos-Yulin Urban Agglomeration, the Central Yunnan Urban Agglomeration, the Central Guizhou Urban Agglomeration, the Lanzhou-Xining Urban Agglomeration, and the Ningxia Yellow River Urban Agglomeration. In this respect, we divided

2016年,全国共3809座WWTPs

all WWTPs into 19 groups based on 19 urban agglomerations and one group for WWTPs located outside urban agglomerations.

Overall, 64 types of wastewater treatment processes are employed in Chinese WWTPs. The 64 types of wastewater treatment processes were classified into nine categories. These categories (Code A to I) are oxidation ditch (OD), anaerobic-oxic (A/O), anaerobic-anoxic-oxic (A^2/O), sequencing batch reactor (SBR), cyclic activated sludge system (CASS), BIOLAK (biological lake; a multi-stage activated sludge process), traditional activated sludge (TAS), biological film (BIOFILM), and others (Table S3). However, the processes of the remaining 99 WWTPs (Code J) are not clear due to lack of information.

On the point of treatment flow (t/day), the WWTPs vary from 最小1400t/日 — 最大:2,800,000t/d 1400 t/day to 2,800,000 t/day. Five intervals were classified into five groups (Code I to V) on the basis of treatment flow, depicted as capacity (shown in Table S4), i.e., super-small capacity ($1400 \le F < 5000$), small capacity ($5000 \le F < 20,000$), medium capacity ($20,000 \le F < 100,000$), large capacity ($100,000 \le F < 500,000$), super-large capacity ($500,000 \le F \le 2,800,000$). In this regard, it should be noted that the treatment flows of 89 WWTPs (Code VI) were unidentifiable.

The WWTP database consists of six properties (i.e., name, location, wastewater treatment process, wastewater treatment flow, wastewater treatment capacity, and DS production) of each WWTP. Of the 3809 WWTPs, 3693 WWTPs have longitude and latitude data and are illustrated in Fig. S1. Among the 3693 WWTPs, 3600 WWTPs have the information of longitude and latitude, wastewater treatment process (Code A to I), and wastewater treatment flow (Code I to V). The following analysis is therefore based on these 3600 WWTPs.

2.3. Sampling strategy

2.3.1. Sampling unit and sampling frame

In summary, the sampling unit indicates each WWTP. From the 3600 WWTPs, 233 WWTPs were of super-small capacity ($1400 \le F < 5000$ t/day) and/or other treatment process. Eighteen WWTPs were of super-large capacity ($500,000 \le F \le 2,800,000$ t/day) and excluded from the sampling frame as they are to be sampled and analyzed distinctively. Thus, the sampling frame in this paper consists of 3349 WWTPs (with treatment process of OD, A/O, A^2/O, SBR, CASS, BIOLAK, TAS, or BIOFILM; with small, medium, and large treatment capacity).

2.3.2. Target indicator and stratification indicators

The ideal target indictor of the survey should be the mass and/or concentrations of targeted chemicals in sludge. However, it has proven to be impossible to obtain the relevant data. Moreover, China has not conducted a national sewage sludge survey and the data relating to the precise concentrations of targeted chemicals present in sewage sludge are unavailable at present. Therefore, the actual target indicator (DS production; the most appropriate and relevant to the ideal target indicator) was used for testing the sampling strategies. As previously mentioned, DS production was calculated from wastewater treatment flow and can be influenced by urban agglomeration, treatment process, and treatment capacity. Urban agglomeration is a comprehensive geographic indicator, representing the production, consumption, and discharge of chemicals in a specific area. Wastewater treatment process is an indicator of all technologies used to remove chemicals during wastewater treatment whilst wastewater treatment capacity is an indicator related to the mass/concentrations of chemicals in influent and/or effluent, as well as sludge. Therefore, urban agglomeration, wastewater treatment process, and wastewater treatment capacity were employed as stratification indicators.

As shown in Table S5, there is only one stratum in Strategy 1 whilst Strategy 2 has 20 strata, divided by the stratification indicator of urban agglomeration. Similarly, Strategy 3 and Strategy 4 have eight strata and three strata, respectively, separated by treatment process and treatment capacity. In Strategy 5, 160 multiple-strata are determined by multiplying 20 (urban agglomerations) and eight (treatment processes). Strategy 6 has 60 strata by a combination of two indicators (urban agglomeration and treatment process). Strategy 7 has 24 strata by treatment process and treatment capacity. Comparatively, Strategy 8 has 480 strata under the interaction among three indicators (urban agglomeration, treatment process, and treatment capacity).

2.3.3. Sample size

It is common knowledge and widely accepted that the larger the sample size, the lesser the sampling error/s being present or introduced into a particular study. However, large sample size has the potential to increase the sampling cost considerably. Thus, a balance between sampling error/s and sampling cost must be reached in order to design an optimal sampling strategy.

The initial sample size (n_0) for all strategies can be calculated using Eq. (1) (Jin et al., 2015) and equals 66.3.

$$n_0 = \frac{N z_{\alpha/2}^2 S^2}{N d^2 + z_{\alpha/2}^2 S^2} \tag{1}$$

where N is the WWTP number in the sampling frame ($N = 3349$); α is the significance level and assumed as 0.10 (i.e., the confidence level 1-α, is 0.90), thus the fractile $z_{\alpha/2}$ (i.e., $z_{0.05}$) is 1.65. Assuming that the detection probability for a specific chemical in sludge follows the binomial distribution, and the detection frequency (P) of targeted chemical in sludge is 50%, thus the variance ($S^2 = P(1-P)$) equals 0.25, maximizing the sample size; d is the error of the detection frequency and assumed as 10% (i.e., the detection frequency is in the range of 40%–60%).

The design effect (*deff*) is widely used in survey sampling, defined as the ratio of the variance of an estimator under a specific random sampling to that under simple random sampling. *Deff* is less than one for stratified random sampling. Herein, *deff* was conservatively set as one and then used to calculate the first-adjusted sample size (n_1) using Eq. (2) (Jin et al., 2015) and n_1 is still 66.3.

$$n_1 = n_0 \times deff \tag{2}$$

To further guarantee the sample size in the real sampling campaign, the effective response rate (r) was set as 80%, and was used to calculate the second-adjusted sample size n_2 using Eq. (3) (Jin et al., 2015) and n_2 is 82.9. This was then rounded to 83 as the sample size for Strategy 1.

$$n_2 = \frac{n_1}{r} \tag{3}$$

For Streategy 2–Strategy 8, the sample size in each stratum is proportionally allocated according to the weight of stratum h (W_h), which is represented by the ratio of WWTP number in stratum h (N_h) to the total WWTP number in the sampling frame ($N = 3349$), as shown in Eq. (4).

$$W_h = \frac{N_h}{N} \tag{4}$$

The sample size in stratum h (n_h) of Strategy2–Strategy8 was given by Eq. (5).

$$n_h = ROUND(n_2 \times W_h) \tag{5}$$

where *ROUND* is to round off an amount to the nearest whole number.

The sample sizes for Strategy2–Strategy8 (n) were calculated as 82, 82, 82, 70, 79, 83, and 53, respectively, by Eq. (6).

$$n = \sum n_h \tag{6}$$

2.3.4. Simulation

A *Monte Carlo* simulation was performed to examine the sampling campaign 10,000 times by *MATLAB* (*R2018a*; *Mathworks*). Then, the mean DS production of the selected WWTPs in stratum h for simulation i ($\bar{x}_{h,i}$) was adopted to extrapolate the mean DS production of the total 3349 WWTPs for simulation i ($\bar{x}_i$) using Eq. (7) (Zhang, 2007).

$$\bar{x}_i = \sum W_h \bar{x}_{h,i} \tag{7}$$

The estimated total DS production of 3349 WWTPs for simulation I ($\hat{x}_i$)x, is given by Eq. (8).

$$\hat{x}_i = \bar{x}_i \times N \tag{8}$$

The root mean square error (RMSE) was then calculated using Eq. (9).

$$\mathrm{RMSE} = \sqrt{\frac{1}{10,000}\sum_{i=1}^{10,000}(x_i - \hat{x}_i)^2} \tag{9}$$

where x_i denotes the true total DS production of 3349 WWTPs (6.06 million t/year) and $\hat{x}_i$ denotes the estimated total DS production. The eight RMSEs for the eight sampling strategies are summarized in Table S5.

3. Results and discussion

3.1. Geographic distribution of Chinese WWTPs

Fig. 1a and d show the density and distribution of 3600 WWTPs in China. It is evident that the Yangtze Delta Urban Agglomeration (around Shanghai) has the highest number of WWTPs (614), followed by the Beijing-Tianjin-Hebei Urban Agglomeration (around Beijing and Tianjin; 306), the Middle Yangtze River Urban Agglomeration (267), the Shandong Peninsula Urban Agglomeration (266), and the Pearl River Delta Urban Agglomeration (around Guangzhou; 224). The Hu Line, also named as the Heihe-Tengchong Line, was discovered by population geographer Hu Huanyong in 1935, and divides the area of China into two approximately equal parts, marking the striking difference in the distribution of the country's population. Roughly 94% of China's population live to the east of the line drawn between Heihe in the north and Tengchong in the south. Similarly, nearly 85% of China's WWTPs are located in the east area of the line.

The distributions of population, GDP, WWTP number, wastewater treatment flow, and DS production of urban agglomerations are generally in good agreement, as shown in Fig. 1b–1f (details listed in Table S6). In 2017, the population of the Yangtze Delta Urban Agglomeration, the Middle Yangtze River Urban Agglomeration, the Beijing-Tianjin-Hebei Urban Agglomeration, and the Shandong Peninsula Urban Agglomeration, reached 152.7 million, 129.4 million, 112.1 million, and 100.1 million, respectively. In terms of GDP, the top four are the Yangtze Delta Urban Agglomeration (16.5 trillion RMB), and the Beijing-Tianjin-Hebei Urban Agglomeration (8.1 trillion RMB), the Middle Yangtze River Urban Agglomeration (7.9 trillion RMB), and the Pearl River Delta Urban Agglomeration (7.6 trillion RMB). As anticipated, the Yangtze Delta Urban Agglomeration treated the largest flow of wastewater (12,507 million t) and dry sludge (1.7 million t) in China in 2017, accounting for approximately 21.0% and 24.1% of the total (59,600.5 million t wastewater and 7.0 million t DS), respectively. This was followed by the Pearl River Delta Urban Agglomeration, the Beijing-Tianjin-Hebei Urban Agglomeration, the Middle Yangtze River Urban Agglomeration, the Shandong Peninsula Urban Agglomeration, and the Chengdu-Chongqing Urban Agglomeration, which contributed to 10.1% (of the total wastewater treatment flow) and 6.8% (of the total DS production), 9.2% and 11.8%, 7.7% and 5.0%, 7.6% and 10.1%, and 4.6% and 4.4%, respectively. According to the *Outline of the Development Plan for the Yangtze River Economic Belt* of China, the Yangtze River Economic Belt consists of the Yangtze Delta Urban Agglomeration, the Middle Yangtze River Urban Agglomeration, and the Chengdu-Chongqing Urban Agglomeration. The population, GDP, WWTP number, wastewater treatment flow, and DS production of the *Belt* account for about one-third of China, i.e., 27.0%, 35.8%, 29.4%, 33.2%, and 33.5%, respectively.

The correlation between population, GDP, WWTP number, wastewater treatment flow, and DS production in each urban agglomeration were further examined. As Table S7 shows, all correlation coefficients are higher than 0.80 with $p < 0.01$, indicating a strong correlation among them.

By comparison, as Fig. S2 shows, the Pearl River Delta Urban Agglomeration treats more wastewater (97.5 t/person/year) than those in other agglomerations, e.g., 89.7 t/person/year and 81.9 t/person/year in the Northern Tianshan Mountain Urban Agglomeration and the Yangtze Delta Urban Agglomeration, respectively. Interestingly, the Northern Tianshan Mountain Urban Agglomeration ranked first for DS production (14.2 kg/person/year), followed by the Yangtze Delta Urban Agglomeration (11.1 kg/person/year) and the Ningxia Yellow River Urban Agglomeration (10.7 kg/person/year). It is evident from this that each of these three productions are more than double the Chinese DS production of 5.1 kg/person/year, which is slightly higher than the previously reported Chinese production of 4.6 kg/person/year (Yang et al., 2015) and lower than the production of European countries (17.7 kg/person/year) (Kelessidis and Stasinakis, 2012).

3.2. Wastewater treatment process and treatment capacity

Ten types of treatment processes were employed in China's WWTPs. In the investigated 3600 WWTPs, nine of them were reported in Fig. 2a, including OD with a percentage of 28.7%, A^2/O (28.1%), CASS (12.8%), A/O (7.3%), SBR (6.9%), BIOFILM (4.4%), TAS (3.3%), BIOLAK (2.9%), and other treatment process (5.6%). The percentages are different from those reported previously, i.e. A^2/O (31%), OD (21%), TAS (11%), SBR (10%), A/O (8%), and BIOFILM (4%) (Zhang et al., 2016). Comparatively, the percentage of OD gets larger, whilst the percentages of A^2/O, SBR, and TAS decrease. Particularly, the percentage of TAS is halved in this instance, probably as a result of TAS being replaced by OD, which covers a small area and could remove nitrogen and phosphorus efficiently, thus meeting the market requirement. These processes in Chinese WWTPs, with COD removal efficiency of about 85% or even higher, could well remove COD and had no significant difference (Jin et al., 2014).

For treatment capacity, the investigated WWTPs were divided into six groups. Five of them were investigated, being super-small WWTPs (1.3%), small WWTPs (32.7%), medium WWTPs (54.3%), large WWTPs (11.2%), and super-large WWTPs (0.5%), as illustrated in Fig. 2b. Jin et al. (2014) reported that large WWTPs have the largest COD removal efficiency (87.5%), followed by super-large WWTPs (86.5%), medium WWTPs (85.5%), and small WWTPs (81%), respectively.

3.3. Monte Carlo simulation of eight sampling strategies

Taking Strategy 1 as an example, a sample (a random set of 82 WWTPs) was taken within the sampling frame 10,000 times and the total DS productions (expressed as 10,000 datapoints in Fig. S3a) of 3349 WWTPs in the sampling frame were estimated. Fig. S3b illustrates the relative frequency distribution of the 10,000 datapoints and the corresponding Gauss fitting curve. The red dashed lines denote the true total DS production (6.06 million t/year). Fig. 3 shows the fitting curves of the eight strategies (all $R^2 > 0.97$) and the true total DS production (6.06 million t/year) in the red dashed line.

In addition, the term "bias" is used here to indicate the difference between the mean (or median) of all estimated total DS productions and the true total DS production, expressed as the mean (or median)

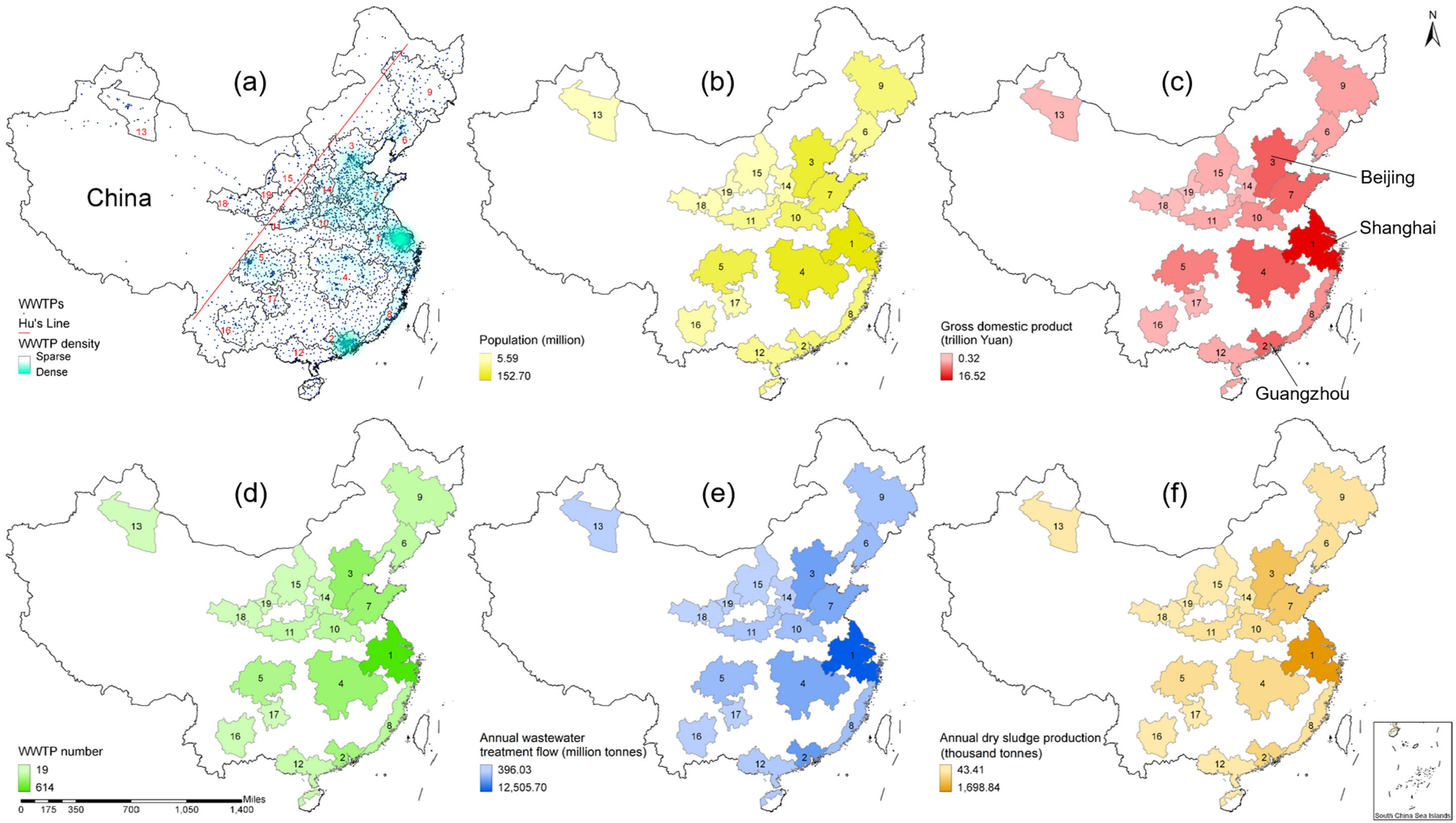

Fig. 1. (a) Distribution of Chinese 3600 WWTPs, (b) population, (c) gross domestic product (GDP), (d) WWTP number, (e) annual treatment flow, and (f) annual dry sludge (DS) production in each urban agglomeration. 1. the Yangtze Delta Urban Agglomeration, 2. the Pearl River Delta Urban Agglomeration, 3. the Beijing-Tianjin-Hebei Urban Agglomeration, 4. the Middle Yangtze River Urban Agglomeration, 5. the Chengdu-Chongqing Urban Agglomeration, 6. the Middle and South Liaoning Urban Agglomeration, 7. the Shandong Peninsula Urban Agglomeration, 8. the West of Taiwan Strait Urban Agglomeration, 9. the Harbin-Changchun Urban Agglomeration, 10. the Central Plain Urban Agglomeration, 11. the Central Shaanxi Plain Urban Agglomeration, 12. the Beibu Gulf Urban Agglomeration, 13. the Northern Tianshan Mountain Urban Agglomeration, 14. the Central Shanxi Urban Agglomeration, 15. the Hohhot-Baotou-Erdos-Yulin Urban Agglomeration, 16. the Central Yunnan Urban Agglomeration, 17. the Central Guizhou Urban Agglomeration, 18. the Lanzhou-Xining Urban Agglomeration, and 19. the Ningxia Yellow River Urban Agglomeration. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Data sources: the *Statistical Book* of 31 provinces, autonomous regions, and municipalities; the 2018 *Urban Drainage Statistical Yearbook* (China Urban Water Supply and Drainage Association, 2019); the 2016 *Development Report on China's Urban Agglomeration* (Fang et al., 2016).

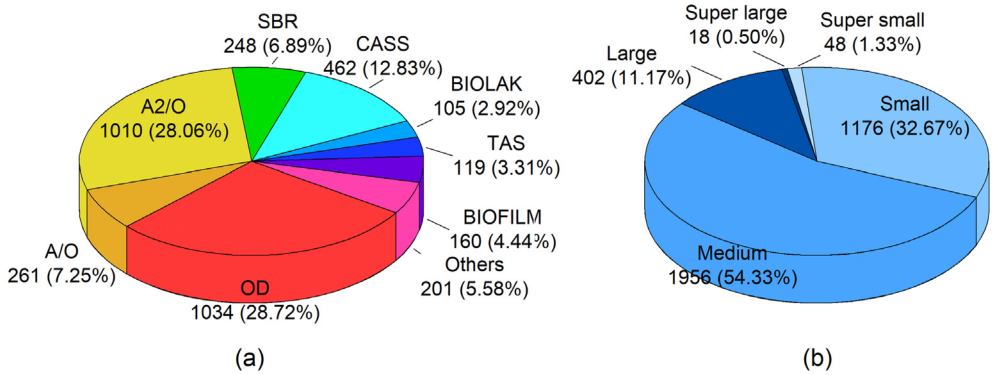

Fig. 2. Distribution of Chinese 3600 WWTPs by (a) wastewater treatment process and (b) wastewater treatment capacity.

relative error (MRE). Furthermore, the term "precision" is used to clarify the variation among all the estimated data, expressed as the standard deviation (SD). In light of this, "accuracy" is a term that combines both the concept of bias and precision, quantifying the difference between the estimated values and the true value (Walther and Moore, 2005), expressed as the root mean square error (RMSE). It is therefore apparent that RMSE is a better and preferred method of choosing a good strategy than solely opting for MRE or SD. All MREs, SDs, and RMSEs are summarized in Table S5.

As Table S5 and Fig. 3 show, compared to Strategy 1 (simple random sampling), no significant improvement on the precision is observed for both Strategy 2 (stratified random sampling by urban agglomeration) and Strategy 3 (stratified random sampling by treatment process). Moreover, Strategy 5 (stratified random sampling by urban agglomeration and treatment process), Strategy 6 (stratified random sampling by urban agglomeration and treatment capacity), Strategy 7 (stratified random sampling by treatment process and treatment capacity), and Strategy 8 (stratified random sampling by urban agglomeration, treatment process, and treatment capacity) introduce higher precision but larger bias. Strategy 4 (stratified random sampling by treatment capacity) presents the smallest RMSE among all strategies and provides higher precision and lower bias when compared with Strategy 1. By multiple stratifications, Strategy 5, Strategy 6, Strategy 7, and Strategy 8 get more precise results. In particular, Strategy 8 with 480 strata, by the use of three stratification indicators, reaches the highest precision in all strategies. However, excessive stratification leads to large bias, which will underestimate the total DS production.

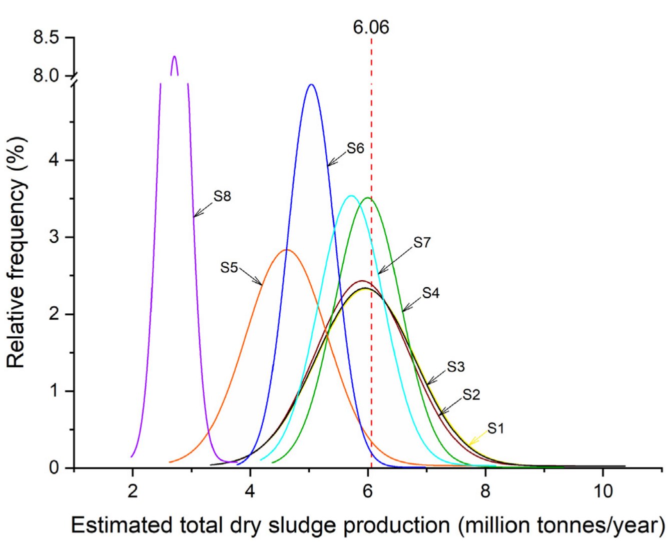

Fig. 3. Gauss fitting curves for the relative frequency distribution of the 10,000 estimated total dry sludge (DS) productions in Strategy 1–Strategy 8. It is noted that the curves pointed by Strategy 1 and Strategy 3 almost overlap but are distinct from each other. Data sources: the 2018 Urban Drainage Statistical Yearbook (China Urban Water Supply and Drainage Association, 2019).

In light of bias, it was found that urban agglomeration was the worst stratification indicator whilst treatment process was slightly better, and treatment capacity was the best. It was determined that any combination of these stratification indicators will have a negative effect on bias. Thus Strategy 8 (the combination of urban agglomeration, treatment process, and treatment capacity) is the worst, followed by Strategy 5 (the combination of urban agglomeration and treatment process), Strategy 6 (the combination of urban agglomeration and treatment capacity), and Strategy 7 (the combination of treatment process and treatment capacity), in descending order of bias.

The sample sizes for the eight strategies above were decided by considering the population, confidence interval, detection frequency, error, design effect, as well as effective response rate. In addition, sample sizes were assumed at 50, 100, 150, 300, 500, 1000, 1500, and 3000, and simple random sampling was conducted 10, 000 times for each sample size. The means, medians, SDs, and RMSEs of estimated total DS productions for each sample size as well as Strategy 1 and Strategy 4 are shown in Fig. S4. With the growth of the sample size, the mean and median increase slowly and tend to a fixed value. By contrast, SD and RMSE decrease. It is apparent that the RMSE of Strategy 4 (sample size: 82) is between the simple random sampling RMSEs (sample size: 150 and 300), indicating that the sampling efficiency of Strategy 4 is high.

3.4. Sample allocation in Strategy 4

For Strategy 4, the sample size is 82 (Table 1), including nine large WWTPs, 46 medium WWTPs, and 27 small WWTPs. In addition, there are six, five, four, two, and one super-large WWTPs existent in the Yangtze Delta Urban Agglomeration, the Pearl River Delta Urban Agglomeration, the Beijing-Tianjin-Hebei Urban Agglomeration, the Chengdu-Chongqing Urban Agglomeration, and the Middle and South Liaoning Urban Agglomeration, respectively. These should be considered in the sampling campaign. On the contrary, no super-small WWTPs were selected. The main WWTPs selected are located in the Yangtze Delta Urban Agglomeration (22), the Pearl River Delta Urban Agglomeration (8), the Beijing-Tianjin-Hebei Urban Agglomeration (15), and the Middle Yangtze River Urban Agglomeration (9) and the Chengdu-Chongqing Urban Agglomeration (6). It should be noted that although the 18 super-large WWTPs are sampled together with the 82 selected ones, they will be further studied independently.

Stratified sampling was successfully applied in the Targeted National Sewage Sludge Survey (TNSSS) by the U.S. EPA in 2006–2007. The stratification employed by the U.S. EPA was by treatment flow which is similar to treatment capacity (the optimal stratification indicator) employed in this study. The U.S. EPA selected 74 WWTPs from a pool of 3337 WWTPs (United States Environmental Protection Agency, 2006) whilst this study sampled 82 WWTPs from 3349 WWTPs using Strategy 4. Moreover, there were more parameters as targets for testing

Table 1
A sample (82 WWTPs) selected by Strategy 4 as well as the 18 super-large-capacity WWTPs.

Name of urban agglomeration	Super-large capacity	Large capacity	Medium capacity	Small capacity	Super-small capacity	Total
Yangtze Delta Urban Agglomeration	6 (6)[a]	69 (2)	299 (9)	224 (5)	16 (0)	614 (22)
Pearl River Delta Urban Agglomeration	5 (5)	49 (1)	127 (2)	38 (0)	5 (0)	224 (8)
Beijing-Tianjin-Hebei Urban Agglomeration	4 (4)	27 (1)	188 (8)	82 (2)	5 (0)	306 (15)
Middle Yangtze River Urban Agglomeration	0 (0)	36 (3)	158 (4)	72 (2)	1 (0)	267 (9)
Chengdu-Chongqing Urban Agglomeration	2 (2)	14 (1)	98 (3)	61 (0)	4 (0)	179 (6)
Middle and South Liaoning Urban Agglomeration	1 (1)	23 (0)	68 (0)	17 (0)	0 (0)	109 (1)
Shandong Peninsula Urban Agglomeration	0 (0)	30 (0)	199 (2)	36 (0)	1 (0)	266 (2)
West of Taiwan Strait Urban Agglomeration	0 (0)	19 (0)	72 (0)	37 (0)	5 (0)	133 (0)
Harbin-Changchun Urban Agglomeration	0 (0)	23 (0)	52 (2)	22 (1)	0 (0)	97 (3)
Central Plain Urban Agglomeration	0 (0)	17 (0)	92 (5)	17 (0)	0 (0)	126 (5)
Central Shaanxi Plain Urban Agglomeration	0 (0)	12 (0)	41 (3)	38 (1)	0 (0)	91 (4)
Beibu Gulf Urban Agglomeration	0 (0)	8 (0)	37 (0)	23 (1)	1 (0)	69 (1)
Northern Tianshan Mountain Urban Agglomeration	0 (0)	7 (0)	16 (1)	10 (1)	0 (0)	33 (2)
Central Shanxi Urban Agglomeration	0 (0)	4 (0)	21 (1)	40 (1)	1 (0)	66 (2)
Hohhot-Baotou-Erdos-Yulin Urban Agglomeration	0 (0)	3 (0)	20 (0)	15 (0)	0 (0)	38 (0)
Central Yunnan Urban Agglomeration	0 (0)	7 (0)	22 (0)	21 (1)	0 (0)	50 (1)
Central Guizhou Urban Agglomeration	0 (0)	4 (0)	12 (0)	19 (1)	2 (0)	37 (1)
Lanzhou-Xining Urban Agglomeration	0 (0)	4 (0)	13 (0)	18 (0)	0 (0)	35 (0)
Ningxia Yellow River Urban Agglomeration	0 (0)	4 (0)	11 (0)	4 (0)	0 (0)	19 (0)
Outside of Urban Agglomeration	0 (0)	42 (1)	410 (6)	382 (11)	7 (0)	841 (18)
Total	18 (18)	402 (9)	1956 (46)	1176 (27)	48 (0)	3600 (100)

[a] WWTP numbers of various treatment capacities in each urban agglomeration are shown outside the parentheses, and the sample sizes in each stratum are within the parentheses.

bias and precision of the strategies in the fourth survey of the United States as the United States had conducted three surveys prior to the fourth study, which yielded a considerable amount of sample data. In contrast, this would be the first national sampling survey conducted in China.

3.5. Limitations and implications for future studies

Several limitations of this study have been identified. One such limitation includes the lack of information available in respect of WWTPs. WWTP lists are issued by various government departments and are considerably different from each other, proving difficult to consolidate. This issue is sometimes compounded by the fact that information collected may be incomplete e.g. lack of information on wastewater sources. In addition, a general platform from which interested parties or researchers may collect WWTP information does not exist. As a result, interested parties or researchers resort to collecting information related to WWTPs from multiple platforms. This, once again, results in the risk of incomplete information being obtained. In respect to stratification indicators, there are two identifiable limitations to the study. Firstly, the stratification indicators used in this study are limited due to the lack of information provided by the relevant government departments and available resources. Secondly, the classification of stratification indicators requires further consideration. In respect of the sampling model, there are also two notable limitations. The first of which is that the focus of this study is singular in nature, with its focus being on decreasing sampling error/s. However, sampling cost is also an essential factor which should be taken into account. As such, it is submitted that models involving cost should be introduced to future sampling strategy designs. The second of which is that this paper only applies simple random sampling to each stratum. Other sampling methods such as systematic sampling can also be employed for sampling within strata in subsequent studies.

The main objective and therefore implication of this paper is that it determines the most optimal sampling strategy for decreasing sample error/s up to the hilt. However, sampling design, although a crucial step in conducting a CNSSS, is merely the first in doing so. It is only when the sampling strategy is connected to the subsequent steps of collecting, transporting, conserving and determining sewage sludge, can the data quality and assurance be achieved. Finally, sewage sludge samples from the CNSSS may contribute to the development of a Chinese sewage sludge sample bank (CSSSB) for further research. In this regard, sample analysis data has the potential to serve as the foundation for sampling strategy improvement.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.133998.

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