

Article

Characterization of Disinfection By-Products Levels at an Emergency Surface Water Treatment Plant in a Refugee Settlement in Northern Uganda

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Received: 28 February 2019; Accepted: 25 March 2019; Published: 28 March 2019



Abstract: The reliance on chlorination in humanitarian operations has raised concerns among practitioners about possible health risks associated with disinfection by-products; however, to date, there has not been an evaluation of disinfection by-product (DBP) levels in an emergency water supply intervention. This study aimed to investigate DBP levels at a surface-water treatment plant serving a refugee settlement in northern Uganda using the colorimetric Hach *THM Plus* Method. The plant had two treatment processes: (1) Simultaneous clarification–chlorination (“rapid treatment”); and (2) pre-clarification and chlorination in separate tanks (“standard treatment”). For both standard ($n = 17$) and rapid ($n = 3$) treatment processes, DBP levels in unique parcels of water were tested at 30 min post-chlorination and after 24 h of storage (to simulate what refugees actually consume). DBP levels after 24 h did not exceed the World Health Organization (WHO) guideline limit of 300 ppb equivalent chloroform, either for standard treatment (mean: 85.1 ppb; 95% confidence interval (C.I.): 71.0–99.1 ppb; maximum: 133.7 ppb) or for rapid treatment (mean: 218.0 ppb; 95% C.I.: 151.2–284.8; maximum: 249.0 ppb). Observed DBPs levels do not appear to be problematic with respect to the general population, but may pose sub-chronic exposure risks to specifically vulnerable populations that warrant further investigation.

Keywords: chlorine; chlorination; disinfection by-products; humanitarian; refugee camp; trihalomethanes; water treatment; water quality

1. Introduction

Chlorination is the most widely used method for water treatment in humanitarian emergencies because of its simplicity, low cost, and importantly, the residual protection it provides. In refugee and internally displaced persons (IDP) camps in humanitarian crisis zones, centralized batch chlorination remains the primary approach to treating large quantities of water, while point-of-use (e.g., household) and point-of-distribution (e.g., chlorine dispensers) approaches are also utilized in specific niche roles [1–5]. A number of chlorine products are commonly utilized in humanitarian response including calcium hypochlorite powders (e.g., high-test hypochlorite, HTH), sodium hypochlorite solutions (e.g., bleach), as well as tablets composed of various chlorine compounds such as sodium dichloroisocyanurate (NaDCC) or chlorine dioxide. When these products react with water, hypochlorous acid and hypochlorite ions are formed, which provide the disinfectant effect. The reliance on chlorination in humanitarian operations, including of raw surface waters during the early weeks of rapid-onset emergencies, has raised concerns among practitioners about possible health risks

due to disinfection by-products [6]. Similar concerns have also been raised about the use of various chlorine products in household water treatment programs in developing country settings [7,8].

Disinfection by-products (DBPs) form when hypochlorous acid reacts with natural organic matter (NOM) such as humic and fulvic acids present in natural surface and ground waters [9]. DBPs are chemically stable and will accumulate in treated water in the presence of chlorine and organic precursors. DBP formation increases with residual chlorine concentration, temperature, contact time, pH, and NOM levels [10–12]. DBPs such as trihalomethanes (THMs) may be linked to cancer and non-cancer adverse health effects (i.e., reproductive system) and are therefore subject to maximum allowable concentrations from the WHO and many national regulators [11,13]. The exact role of THMs versus other DBPs (over 600 species have been identified) in causing adverse health outcomes is unclear; THMs are likely a surrogate measure for other more hazardous DBPs [14]. Current epidemiological evidence shows a consistent association between long-term THM exposure (30+ years) and risk of bladder cancer, although the causal nature of the association is not conclusive, whereas the epidemiological evidence concerning other cancer sites is insufficient or mixed [15–21]. Current evidence also suggests minor effects from high THM exposure during pregnancy on fetal growth indices such as small for gestational age at birth, but is inconclusive with respect to other reproductive outcomes (e.g., low birth weight, fetal loss, preterm delivery, and congenital malformation) [15,22,23].

Globally, displacement crises are becoming increasingly intractable, with populations forced to remain in refugee and IDP camps for extended periods of time, sometimes even for decades. As a consequence, health risks associated with chronic exposures, which were previously considered to be less relevant in humanitarian response situations, are becoming increasingly pertinent (see, for example, Reference [24]). DBPs are one such concern that humanitarian responders must start thinking concretely about. To our knowledge, there has not been an evaluation of DBP levels in an emergency water supply intervention before. In response to this gap, we investigated DBP levels using field-appropriate methods in production water at an emergency surface water treatment plant serving a refugee settlement in northern Uganda. In this paper, we describe the investigation undertaken, present findings, and discuss their implications for humanitarian response. In doing so, this paper provides the first characterization of DBPs in drinking water supplies in a humanitarian field setting.

2. Materials and Methods

2.1. Site Background

We carried out our investigation at a large emergency surface-water treatment plant (SWTP) located on the White Nile in northern Uganda (located at: 3°22'38.42" N 31°38'24.42" E). The plant was built in early 2017 by *Médecins Sans Frontières* (Operational Centre Amsterdam, OCA) who continued to operate the system at the time of the study in August and September 2017 (it would eventually be handed over to the Ugandan Red Cross in early 2018). The SWTP could, at the time of the study, produce over 1200 m³/day of treated drinking water, which supplied the nearby Palorinya refugee settlement (population: 183,700 at the time of the study) with a large part of its total water needs (Figure 1).



Figure 1. Image of multiple clarification tanks, clear tanks, and pumping to tanker trucks at the Palorinya surface-water treatment plant (SWTP), August 2017 (Source: Syed Imran Ali).

The treatment processes implemented at the Palorinya SWTP changed over time reflecting the typical evolution of emergency water supply in terms of quantity and quality:

- Initially, during the acute phase of the emergency, overall quantity was emphasized. Raw river water was directly chlorinated in tanker trucks in order to maximize quantities and speed of delivery to the population.
- Next, in order to improve quality, temporary clarification tanks were installed in which water was simultaneously clarified and chlorinated with aluminum sulphate and high-test hypochlorite (HTH). The simultaneous oxidation step was done to help control NOM content as well as an early operating issue with floating flocs in the settling tanks. Following dosing, waters were allowed to clarify and react for a short time before being pumped to trucks for delivery to the settlement (“rapid treatment”).
- Finally, in order to further reduce turbidity and improve chlorine residual stability, the process was upgraded to “standard” two-stage treatment with pre-clarification using polyaluminum chloride (PAC) in sedimentation tanks followed by chlorination using HTH in separate disinfection tanks with adequate contact time before being pumped to trucks for delivery to the settlement (“standard treatment”).

At the time of the study, the SWTP was in the process of being upgraded so one half of the plant still utilized simultaneous clarification–chlorination (“rapid treatment”) and the other half had been upgraded to two-stage treatment (“standard treatment”). The free residual chlorine (FRC) level in production water output from the Palorinya SWTP was set to 1.6 mg/L in order to ensure sufficient residual for truck transport, storage in local water tanks in the settlement (observed mean FRC: 0.7 mg/L), and storage in refugees’ households (observed mean FRC: 0.4 mg/L).

2.2. DBP/THM Measurement

The laboratory standard for the measurement of DBPs is gas chromatography, which requires specialized equipment that is generally unsuitable for humanitarian field settings [25–28]. We therefore sought to use more field-appropriate methods that could realistically be implemented at this and other humanitarian field sites. We used the colorimetric *THM Plus* Method produced by Hach (Dr. Lange Nederland B.V., Tiel, the Netherlands) (Hach Method #10132). The *THM Plus* Method is demonstrated to have good correlation with instrumental reference methods and has an established track record of

use in small-scale water treatment facilities in the US [29,30]. The method requires relatively simple inputs including hot and cold water baths, four chemical reagents, and a battery-operated Hach DR1900 spectrophotometer (a full explanation of the method and materials is available in the manufacturer's method note: Reference [31]). The *THM Plus* Method measures a total of 11 trihalogenated DBP species including the four total trihalomethane (TTHM) compounds that are regulated under the US Safe Drinking Water Act (SDWA), plus seven other DBPs including some haloacetic acids (HAAs) and other DBP types (Table 1) [30]. The *THM Plus* Method reports the cumulative sum of these 11 compounds in units ppb equivalent chloroform.

Table 1. Eleven disinfection by-products (DBPs) measured by the Hach *THM Plus* Method.

Name of DBP Compound	Type/Group
Trichloromethane (TCM, chloroform)	TTHM
Dibromochloromethane (DCBM)	TTHM
Bromodichloromethane (BDCM)	TTHM
Tribromomethane (TBM, bromoform)	TTHM
Trichloroacetic acid (TCAA)	HAA
Dichlorobromoacetic acid (DBCAA)	HAA
Bromodichloroacetic acid (BDCAA)	HAA
Tribromoacetic acid (TBAA)	HAA
Chloral hydrate (CH)	Haloacetaldehyde
1,1,1-trichloro-2-propanone (111-TCP)	Haloacetone
1,1,1-trichloroacetoneitrile (TCAN)	Haloacetoneitrile

2.3. DBP/THM Guidelines

A number of national and international health-related guidelines stipulate maximum allowable concentrations of DBP/THM compounds in drinking water. We opted to use the WHO guideline as our primary point of reference as it is an internationally recognized standard applicable to water systems in low- and middle-income countries. The WHO guideline for chloroform of 300 ppb was specifically selected as it corresponds with the output units of the Hach *THM Plus* Method (Table 2). It has also been used to evaluate DBP-related health hazards in safe water programs in development settings [7,8]. The US SDWA limits are also reproduced in the table below as the Hach *THM Plus* Method was developed with reference to these guidelines.

Table 2. Summary of health-related DBP/trihalomethanes (THM) guidelines from WHO and US sources.

Source	Year	DBP Compound Regulated	Guideline Value	Notes
WHO Guidelines for Drinking-Water Quality [32]	2008	Chloroform	300 ppb	Previous chloroform guideline was 200 ppb, but has been increased in the latest revision.
		BDCM	60 ppb	
		DBC	100 ppb	
		Bromoform	100 ppb	
		Total THMs	All THM ratios ≤ 1	The TTHM guideline states that no individual TTHM compound should exceed its specific guideline.
US Safe Drinking Water Act [33]	2006	Total THMs	80 ppb	Sum total of all four total trihalomethane compounds must not exceed guideline value.
		HAA5	60 ppb	The HAA5 group includes monochloroacetic acid, dichloroacetic acid, trichloroacetic acid, bromoacetic acid, and dibromoacetic acid. Of these, only trichloroacetic acid is included in the Hach <i>THM Plus</i> method. Its individual maximum contaminant level goal is 0.2 mg/L (200 ppb).

There is an important discrepancy to note between the compounds screened for by the Hach *THM Plus* Method (Table 1) and the specific compounds regulated by the WHO and US SDWA guidelines (Table 2). The *THM Plus* Method reports the sum of the concentration of 11 DBP species in units of ppb

equivalent chloroform; this sum includes the four SDWA-regulated TTHM compounds plus four HAAs (of which only one is part of the SDWA-regulated HAA5 group) plus three other DBPs. (For this reason, we refer to the measurement outputs from the Hach *THM Plus* Method as “DBP/THMs”). The WHO and US SDWA guidelines specify concentration limits for either a specific individual DBP compound, or for the TTHM or HAA5 groups. This discrepancy means that the *THM Plus* Method overreports the compounds that are regulated in the WHO and US SDWA guidelines. No individual DBP or DBP group that the Hach method detects is expected to be greater than the test measurement. On this basis, the Hach *THM Plus* Method might be considered to be inherently conservative. Interestingly, Lantange et al. [7,8] in their study of TTHM production in household chlorination programs in Kenya and Tanzania observed that chloroform was the dominant THM species in the samples they analyzed, contributing on average 78% and 83% of TTHM content across multiple water types in either country, respectively. Similarly, Légaré-Julien et al. [34] also found the speciation of TTHMs to be primarily chloroform when natural surface waters were chlorinated. This reinforces the assumption that the DBP/THM reading produced by the Hach *THM Plus* method can meaningfully be compared to the WHO chloroform guideline as the majority species is indeed likely to be chloroform.

2.4. Data Collection and Analysis

The Hach *THM Plus* Method kit was set up at the on-site water quality laboratory at the Palorinya SWTP (Figure 2). DBP testing was conducted at the SWTP between 22 August 2017 and 19 September 2017. We focused DBP testing on the side of the SWTP with two-stage “standard” treatment, with a limited run of samples conducted on the “rapid” treatment side having simultaneous clarification–chlorination. We followed the evolution of DBPs in chlorinated water by following unique parcels of water and testing water quality at two points: (i) First, at 30 min post-chlorination, and then (ii) again after 24 h of storage in sealed polyethylene terephthalate (PET) bottles (common commercially available plastic bottled water bottles). Stored water was kept in a cool, dry place out of direct sunlight at the on-site water quality laboratory. We tested the same parcel of water after 24 h of storage in order to simulate what users may consume in the refugee settlement. In total, we tested 20 unique parcels of water including 17 from the “standard” treatment side and 3 from the “rapid” treatment side. We compared DBP measurements to the WHO guideline limit for chloroform of 300 ppb in order to assess whether a DBP-related health hazard may exist or not. Raw water from the Nile source was also collected and analysed for key water-quality parameters including turbidity, pH, electrical conductivity, alkalinity, and apparent color as part of normal operations at the SWTP. Data were analysed using STATA 13.1 (StataCorp, College Station, TX, USA) and Microsoft Excel 2011 (Microsoft Inc., Redmond, WA, USA). And raw field data is available as a Supplementary Materials in Microsoft Excel format. As the study did not involve any human participants and only water quality data at the SWTP was collected, the study was exempted from full ethics review by the MSF OCA Medical Director.



Figure 2. Hach *THM Plus* Method kit set up at the on-site Palorinya SWTP water quality lab (Source: Syed Imran Ali).

3. Results

Overall water quality of the raw surface water source (White Nile) treated by the Palorinya SWTP is given in Table 3. An image of the raw water intake of the SWTP is given in Figure 3. Overall, the high turbidity and apparent color of the source water indicates there was likely elevated NOM in the raw water, which would increase the likelihood and extent of DBP generation.

Table 3. Raw water quality of surface water source during August–September 2017 at the Palorinya SWTP (data taken from SWTP monitoring records).

Parameter	Mean (95% C.I.)	Drinking Water Reference Range	Reference Source
Turbidity (NTU)	15.8 (13.2–18.4)	<5	US EPA Drinking Water Standards [35]
pH	7.2 (7.1–7.3)	6.5–8.5	US EPA Drinking Water Standards [35]
Electrical Conductivity ($\mu\text{S}/\text{cm}$)	150.7 (143.4–158.1)	0–800 ^a	US EPA Drinking Water Standards [35]
Alkalinity (mg/L CaCO_3)	66.1 (60.4–71.8)	30–500	Ontario Drinking Water Standards [36] ^b
Apparent Color (units Pt-Co)	133.0 (92.1–174.0)	15	US EPA Drinking Water Standards [35]

^a Based on the US Environmental Protection Agency (EPA) Secondary Drinking Water Regulation for total dissolved solids (TDS) of 500 mg/L. ^b No reference range for alkalinity is promulgated by US EPA so the Operational Guideline for the Ontario Drinking Water Standards is used instead.



Figure 3. Raw water intake from the White Nile at the Palorinya SWTP in August 2017; note marshy condition of river (Source: Syed Imran Ali).

DBP/THM findings at the Palorinya SWTP during the August–September 2017 study period are reported in Table 4.

Table 4. DBP/THM levels observed at the Palorinya SWTP following standard and rapid treatment.

Treatment Process	Time Elapsed	Number of Samples	Mean FRC (mg/L) (95% C.I.)	DBP/THM (ppb)				
				Mean	Min	Max	Standard Deviation	95% C.I.
Standard Treatment	30 min	17	1.76 (1.32–2.18)	59.4	36.0	91.0	17.9	50.1–68.6
	24 h	16	0.74 (0.47–1.01)	85.1	44.7	133.7	26.4	71.0–99.1
Rapid Treatment	30 min	3	0.30 (0–1.36)	202.5	96.0	282.3	96.0	n/a *
	24 h	3	0.27 (0–0.87)	218.0	201	249	26.9	151.2–284.8

* 95% C.I. not reported as small sample size ($n = 3$) and large standard deviation.

Overall, we found that DBP/THM levels after 24 h did not exceed the 300 ppb WHO guideline limit, either for standard treatment (mean: 85.1 ppb; 95% C.I.: 71.0–99.1 ppb; maximum: 133.7 ppb) or for rapid treatment (mean: 218.0 ppb; 95% C.I.: 151.2–284.8; maximum: 249.0 ppb). As expected, the rapid treatment side yielded substantially higher DBP/THM levels compared to the standard treatment side. While the maximum observed levels on the rapid treatment side did not exceed the 300 ppb WHO guideline, they did approach it, getting up to the 250 ppb range. Given that the *THM Plus* Method reports the cumulative total of chloroform plus 10 other DBP compounds, the true concentration of regulated chloroform in the sampled waters must be equal or less than the observed readings, reinforcing the assessment that observed readings did not exceed WHO guidelines for chloroform DBP in treated drinking water.

4. Discussion

Overall, our findings did not indicate that a DBP-related health hazard is created when surface water is chlorinated at the Palorinya SWTP vis-à-vis the WHO guideline limit for chloroform. Additionally, our findings show that “standard” two-stage water treatment with clarification and chlorination in separate tanks results in much lower levels of DBPs in production water as compared to “rapid” treatment with simultaneous clarification–chlorination in the same tank. When water is subject to rapid treatment in which chlorination is done before the water is properly clarified, DBP/THM levels in production water will be elevated and may approach or exceed the WHO guideline limit. The elevated levels of DBPs under rapid treatment likely arise due to a combination of greater availability of NOM as well as higher doses of chlorine in order to achieve sufficient residual.

This finding reinforces the need to transition away from rapid water treatment to two-stage treatment as soon as is feasibly possible in emergencies. This is, of course, notwithstanding the fact that ensuring adequate chlorination to protect against waterborne pathogens remains the priority, as the health risks associated with waterborne diseases far outweigh those linked to DBPs, even in industrialized-world settings (and this difference is likely to be even more pronounced in emergency and resource-poor settings) [14,37–39]. Efforts to control the potential health risks associated with DBPs must not compromise waterborne pathogen control.

While we cannot generalize the findings from the Palorinya SWTP to all emergency surface water treatment interventions globally, this study may represent a “worst-case scenario”, as the SWTP water intake was downstream of a slow-moving, highly vegetated, and marshy area of the White Nile. Moreover, the study took place during the rainy season when NOM levels may be elevated due to surface runoff and associated soil erosion (albeit, in the dry season, lower river flow would reduce dilution which would also act to concentrate and elevate NOM levels). Fortunately, even in the case of a marshy surface-water source, effective pre-clarification before chlorination resulted in DBP/THM levels remaining well below guideline limits, even after 24 hours of storage.

The DBP/THM levels we observed at the Palorinya SWTP correspond with levels observed by other workers investigating water chlorination programs for public health protection in developing-country settings. Lantagne et al. [7], in their investigation of point-of-use chlorination programs in Kenya, observed TTHM levels around 150 ppb 24 h after river water (with a substantially higher turbidity than ours at 305 NTU) was directly chlorinated using 1% sodium hypochlorite solution (FRC at 24 h was 0.07–3.64 mg/L, similar to the FRC range we observed as well). This water type is comparable to the “rapid treatment” samples we observed that were directly chlorinated without clarification, which had a mean DBP/THM of 218.0 ppb (Table 4). Interestingly, the more “clear” water types that Lantagne et al. observed in Kenya (with respect to turbidity and organics content), such as rainwater and well water, had TTHM concentrations in the range of 30 to 80 ppb, which corresponds with DBP/THM levels we observed in “standard treatment” samples which were clarified prior to chlorination (mean 85 ppb). Similarly, in point-of-use chlorination programs in Tanzania, Lantagne et al. [8] observed TTHM levels in the range of 100 ppb at 24 h when river water (turbidity: 5.1 NTU) was directly chlorinated with sodium hypochlorite solution (FRC: 0.8–1.2 mg/L at 24 h). In Tanzania, Lantagne et al. observed substantially lower TTHM levels (10 to 60 ppb) when clearer water types such as tap, well, or lake waters were directly chlorinated. Importantly, none of the DBP levels Lantagne et al. observed in either Kenya or Tanzania exceeded the 300 ppb WHO chloroform guideline. Similarly, Nhongo et al. [40] found THMs to generally be within WHO guideline limits in their investigation of DBP levels in the piped water supply system of Harare, Zimbabwe. Légaré-Julien et al. [34] found THM levels in the range of 50 ppb when natural surface waters from Quebec, Canada were treated using a point-of-use coagulant-disinfectant product that included NaDCC as the disinfecting agent. On the other hand, Werner et al. [41] found levels of chloroform and TTHMs approaching and even exceeding the 300 ppb WHO guideline at 24 h when point-of-use chlorine dioxide tablets were used to treat natural surface waters in the Northern British Isles. Although these latter two studies were not

conducted in developing-country settings, the chlorine tablets they evaluated are intended for use in public health promotion programs in developing-country settings.

The DBP levels observed in this and other studies raise an important question regarding the point of reference used to assess whether a health risk exists (or not). The WHO guideline limit we and other workers have used, of 300 ppb chloroform, relates to health concerns associated with chronic exposure (30+ years). As mentioned at the beginning, there are, however, specifically vulnerable subpopulations, pregnant women namely, for whom there may be adverse reproductive effects at sub-chronic levels of exposure (on the order of weeks and months). Current evidence suggests minor effects from high THM exposures (on the order of 100 ppb with respect to TTHM concentration) during pregnancy on fetal growth indices and other reproductive outcomes. [15,22,42]. This is, indeed, within the range we observed at the Palorinya SWTP, as well as that observed by Lantagne et al. in household chlorination programs in Kenya and Tanzania [7,8]. Further investigation of DBP/THM levels in emergency water supplies, especially during the early weeks of a rapid-onset emergency when rapid water-treatment methods may be in use, and the associated risk of adverse reproductive effects for pregnant women warrants further investigation.

As the first report on DBP/THM levels in production water from an emergency water treatment system, this study is a first step toward understanding a water-related health risk that humanitarian responders will likely have to contend with more in coming years. Further field data on DBP levels in emergency water supplies are needed in order to better characterize the extent of the issue. Based on our experience in Uganda, we found the Hach *THM Plus* Method to be a simple, inexpensive, and effective tool for monitoring DBP/THM levels in humanitarian field settings. We were able to effectively implement the method at the Palorinya SWTP to produce credible readings, both with and without supervision once water quality technicians at the on-site water quality lab were trained on how to collect, analyze, and report DBP/THMs using the *THM Plus* kit. Procedurally, the method is not complicated nor labor-intensive, and local staff were comfortable doing it independently after one week of training and supervision. We recommend it for further field use.

Even with a colorimetric method however, continual monitoring of DBP/THMs introduces additional expense and labor demands to routine monitoring which is already prone to gaps in humanitarian operations. As such, in order to provide useful water quality intelligence while minimizing demands on field teams and resources, proxy measurements for DBP/THM production should also be explored. NOM precursors to THMs can be estimated relatively simply in the field by measuring surrogate parameters such as UV₂₅₄ absorbance [43,44]. Field-appropriate proxy methods should be explored further.

Finally, DBP control strategies for humanitarian field settings may need to be explored if the risk to vulnerable sub-populations is determined to be significant enough. Ideally, the formation of DBPs should be prevented through effective pre-clarification before chlorination. If the common pre-clarification methods used in emergency water treatment are found to be insufficient with respect to NOM removal, enhanced coagulation or adsorption with powdered or granular activated carbon (PAC/GAC) could potentially be utilized to further reduce NOM levels [45–47]. Ceramic nanofiltration membranes have also been evaluated for NOM precursor removal but are technically complex and energy intensive to operate [48]. Once DBPs are formed, they may be removed by multiple methods including aeration techniques such as diffused-air systems or aeration towers (these may not be suitable for humanitarian field settings however); adsorption using PAC or GAC, or nitrifying bio-filters (slow sand filters) [45,49]. Such systems would need to be adapted and evaluated for humanitarian field use.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2073-4441/11/4/647/s1>, raw field data is available as a supplementary file in Microsoft Excel format.

Author Contributions: Conceptualization, S.I.A., M.A., and J.-F.F.; methodology, S.I.A., M.A., F.L., and J.-F.F.; formal analysis, S.I.A.; investigation, S.I.A., M.A., and F.L.; resources, M.A. and J.-F.F.; data curation, S.I.A.; writing—original draft preparation, S.I.A.; writing—review and editing, S.I.A., M.A., F.L., and J.-F.F.; visualization, S.I.A.; supervision, S.I.A., M.A., and J.-F.F.; project administration, M.A., J.-F.F., and S.I.A.; funding acquisition, M.A.

Funding: This research received no external funding.

Acknowledgments: We would like to kindly acknowledge Abukoji Godfrey and Akuku Mawa Nelson for their excellent water quality laboratory work at the Palorinya SWTP including collecting data for this study. In addition, we are greatly for the excellent support of the Amsterdam Procurement Unit (APU) and the Emergency Desk (E-Desk) of *Médecins Sans Frontières* Operational Centre Amsterdam (MSF-OCA).

Conflicts of Interest: The authors declare no conflict of interest.

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