water

MDPI

Article

The Characteristics and Traceability Analysis of the Overflow Pollution During the Flood Season in an Urban Area

Shaofeng Yan [1,2,†], Hongbin Xu [3,†], Yingke Fang [3,*], Jie Li [1], Mingzhe Lv [3], Guoqiang Li [3], Long Huang [3], Yuan Li [3] and Gangfu Song [2]

1 Henan Provincial Technical Center for Ecology and Environment, Zhengzhou 450046, China; shaofengyan@126.com (S.Y.); hjbzwyh@163.com (J.L.)
2 School of Water Conservancy, North China University of Water Resources and Electric Power, Zhengzhou 450046, China; sgf@ncwu.edu.cn
3 School of Ecology and Environment, Zhengzhou University, Zhengzhou 450001, China; xuhongbin_gy@zzu.edu.cn (H.X.); lvmingzhe0609@126.com (M.L.); liguoqiang@zzu.edu.cn (G.L.); hl2130@126.com (L.H.); liyuan7626@zzu.edu.cn (Y.L.)
* Correspondence: fangyingke@zzu.edu.cn; Tel.: +86-17812276984
† These authors contributed equally to this work.

Abstract: The issue of combined sewer overflow (CSO) triggered by rainfall has become a significant obstacle to the improvement of water environment quality. This study conducted a long-term monitoring of three types of rainwater outlets, i.e., combined sewer overflows (Test-CSO), separated sewer outlets (Test-SSO), and partially separated sewer outlets (Test-PSSO), to reveal the characteristics of overflow pollution and trace its sources by monitoring the pollutants from different underlying surfaces across various urban functional areas. The results showed that the major pollutants in overflow events exhibited the following order: COD $\geq$ TSS > TN > TAN > TP. Rainwater elevated COD and TSS in the Test-CSO, while reducing nitrogen and phosphorus concentrations by dilution. The Test-PSSO experienced varying degrees of overflow pollution, primarily due to the sewer sediment. A negative relationship between the rainfall and peak time of overflow pollution was observed. The traceability analysis indicated the overall pollution intensity exhibited the following order: residential areas > industrial parks > commercial areas. In addition to commercial areas, the pollution intensity across underlying surfaces generally exhibited the following order: roofs > roads > grasslands. The roof runoff was an important source of pollutants for overflow pollution, and TSS and COD were the major contributors. Notably, grasslands had a buffering effect on pollutants and pH.

Keywords: overflow pollution; combined sewer; underlying surface; rainfall; first flush

check for updates

Citation: Yan, S.; Xu, H.; Fang, Y.; Li, J.; Lv, M.; Li, G.; Huang, L.; Li, Y.; Song, G. The Characteristics and Traceability Analysis of the Overflow Pollution During the Flood Season in an Urban Area. *Water* **2024**, *16*, 3159. https://doi.org/10.3390/w16223159

Academic Editor: Craig Allan

Received: 8 October 2024
Revised: 24 October 2024
Accepted: 2 November 2024
Published: 5 November 2024

1. Introduction

In recent years, the quality of water environments in most regions of China has been improved significantly. However, non-point source pollution remains a major challenge, particularly overflow pollution during the flood season. This type of pollution is difficult to control due to its dispersed, hidden, random, and cumulative nature. Challenges in laying out monitoring points and quantifying pollution intensity in the flood season make it a significant barrier to further water quality improvements. In some urban areas, there is a notable problem of pollution accumulating during dry seasons and being discharged in bulk during the rainy seasons. This issue is common in older districts with combined sewer systems or incomplete separated sewer systems. This situation is a major cause of water quality deterioration of urban river.

During the rainfall in flood seasons, large amounts of rainwater enter drainage pipes, often exceeding the pipes' design capacity. Consequently, sewage is discharged into urban water bodies through overflow, causing serious pollution [1]. In areas with combined sewers, the intensity of this pollution is magnified, a phenomenon known as combined sewer overflows (CSOs) [2]. Overflow sewage contains complex pollutants, including domestic sewage, rainwater, and sediments from drainage pipes [3–5]. These pollutants consist of organic matter, suspended solids, nitrogen, phosphorus, heavy metals, pesticides, hydrocarbons, pathogens, etc. [6–8]. Upon entering urban water bodies, organic matter rapidly depletes dissolved oxygen, negatively affecting the survival and growth of the aquatic life [9]. Nitrogen and phosphorus lead to eutrophication, causing harmful algal blooms and even functional loss of water bodies [10]. High concentrations of suspended solids accelerate water darkening and odorization [5]. Additionally, pathogens in the overflow sewage pose threats to public health [11].

The intensity of overflow pollution is influenced by the characteristics of water quality and is closely related to the functional characteristics of the urban area [12,13]. For example, overflow pollution differs significantly between residential and industrial areas. Overflow pollution's duration and intensity are also affected by rainfall parameters [14]. Factors such as rainfall volume, duration, and the characteristics of underlying surfaces alter the nature of the rainwater entering drainage pipes, thereby affecting the process of the overflow pollution [15,16].

In the present study, long-term monitoring of pollutants from rainwater outlets under different rainfalls and pollutant concentrations in the rainwater from various underlying surfaces were conducted. Using correlation analysis, the study revealed the characteristics of overflow pollution during the flood season and explored the role of the rainwater in the overflow pollution process, aiming to provide a theoretical basis for controlling overflow pollution.

2. Materials and Methods

2.1. Sample Collection from Rainwater Outlets

This study was conducted in the old urban areas of Zhengzhou, Henan Province, China (E 113°41′–113°46′, N 34°39′–34°57′). Zhengzhou has a temperate continental monsoon climate with distinct seasons, an average annual temperature of 15.4 °C, and an annual rainfall of 631.3 mm. The period of May to September is the flood season of Zhengzhou, and the July and August are the major stages. Three rainwater outlets were selected as long-term sampling points: a combined sewer outlet (Test-CSO), a separated sewer outlet (Test-SSO), and a partially separated sewer outlet (Test-PSSO) that irregularly discharges sewage on dry days (Figure 1). From July 2023 to August 2024, the effluent in 14 rainfall events was sampled, with the rainfalls ranged from 1.1 mm to 114 mm (Table S1).

Long-term sampling was carried out using an automatic sampler (LHMCT-2018, Lihero, Changsha, China; Figure 1). A 30 cm high removable baffle was installed at the outlets to facilitate sample collection. The automatic sampler was equipped with a rainfall-triggered device that activated the sampling program when the rainfall reached 0.4 mm. The triggering parameter was determined according to the minimum threshold required to form runoff. The sampler collected water samples every 10 min. This was later was adjusted to 15-min intervals with a 2-min sampling period for longer duration rains. Sampling continued until the rainfall fell below 0.4 mm or 12 samples were collected. Each water sample consisted of 500 mL of sewage, following an initial 200 mL flush. Samples were kept at 4 °C and transported to the laboratory for the further analysis.

Figure 1. The positional information of the sewer outlets and the equipment for the sampling. (**a**) Study area (data from the Google Map); (**b**) autosampler; (**c**) sewer outlet; (**d**) rainfall monitoring device.

2.2. Sample Collection from Different Underlying Surfaces

To investigate rainwater's role in overflow pollution, rainwater samples were collected from various urban functional areas, specifically from industrial parks, residential areas, and commercial areas. Rainwater was sampled from different underlying surfaces in these areas, i.e., roofs, grasslands, and roads. Sampling processes began once runoff was formed, with a 10-min interval between collections. Samples were stored at 4 °C until laboratory testing.

2.3. Water Quality Testing

Water samples were analyzed for chemical oxygen demand (COD), total ammonia nitrogen (TAN), total nitrogen (TN), total phosphorus (TP), and total suspended solids (TSS) using standard methods published by the American Public Health Association (APHA) [17]. For COD, samples were digested at 150 °C for 2 h after the addition of potassium dichromate and other reagents, and absorbance was measured at 420 nm. Nitrogen and phosphorus concentrations were determined using a spectrophotometer (DR6000, HACH, Loveland, CO, USA) after color development, with absorbance readings taken at 420 nm, 220 nm, 275 nm, and 700 nm, depending on the indicator. Total suspended solids were measured by filtration, drying, and weighing. pH was measured using a pH meter (PHS-3C, Lei Ci, Shanghai, China).

3. Results and Discussion

3.1. Characteristics of the Overflow Pollution

The rainfall events at the three sampling points were not identical due to their different locations. For instance, one point may meet the collection threshold while another may not experience rainfall. To ensure data comparability, water quality data were classified and analyzed to identify the characteristics of the overflow pollution at the three types of rainwater outlets.

3.1.1. Water Quality Characteristics of the Three Types of Outlets

A complete short-time rainfall event (50 min, 3.8 mm) was used to analyze the overall water quality characteristics of the three target outlets (Figure 2). Among the three outlets, the Test-SSO exhibited the lowest concentrations of COD, TAN, TN, TSS, and TP. Throughout the entire rainfall event, no significant fluctuations were observed, indicating no obvious overflow pollution. The primary source of pollution may be rainwater rather than residual pollutants washed out from its pipeline. The following order of pollutant concentrations was observed: COD > TSS > TN > TAN > TP. These results suggested that the efforts to remold combined systems into separate systems were regarded as a thorough and effective method to control overflow pollution [18].

In contrast, the TEST-CSO had the highest concentrations of nitrogen and phosphorus pollutants, which decreased over time as rainwater continuously diluted the sewage. Nitrogen and phosphorus in domestic sewage were the main sources of inorganic pollution, with ammonia nitrogen as the dominant nitrogen form. COD and TSS showed an initial increase followed by a decrease, peaking at 40 and 30 min, respectively. This suggests that rainwater exacerbated the overflow pollution, particularly for COD and TSS. The outlet had similar pollutant types as observed at Test-SSO. Similar results were observed in the study by Chen et al. [19], in which the concentration of particulate pollutants increased sharply in a short time, while the concentration of dissolved pollutants decreased at a certain dilution. The phenomenon of “sharp increase in a short time” is known as the first flush effect [20]. However, the study by Niazkar et al. [20] demonstrated that the occurrence of the first flush could not be unanimously confirmed by all types of pollutants, and it could occur for various types of pollutants in different rainfall events, which was consistent with the results of the present study.

The Test-PSSO displayed significant first flush overflow pollution for TAN, total TN, and TP, with peaks at 10 min. This outlet discharged intermittently during the dry season, retaining large amounts of pollutants in the pipeline, which were later flushed out by rainwater in the wet season. The first flush effect was regarded as an important source of overflow pollution [21]. In the first flush effect, the first 30% of runoff should carry at least 80% of the pollution load during a rainfall [22]. The trend of these pollutants’ concentrations indicated that nitrogen and phosphorus pollution mainly originated from residual pipeline pollutants. Importantly, COD and TSS concentrations increased after 10 min and continued to rise even after the rain stopped. Li et al. [18] reported that the peak pollutant concentrations occurred 40–60 min after the overflow began in the separate stormwater systems inappropriately connected with sewage. The peak time was closely related to the rainfall intensity. The results suggested that the impact of overflow pollution can even last very long periods. Notably, the concentrations of COD and TSS were even higher than that of Test-CSO. Similar results were observed in the study by Yin et al. [23], in which the overflow concentrations of storm drains with inappropriate sewage entry were close to or even higher than that of combined sewers under wet weather period, due to the serious sediment accumulation in the pipes. This suggests that at Test-PSSO, TSS and COD were the dominant pollutants, but their overflow occurred later, with longer durations and higher intensities than nitrogen and phosphorus.

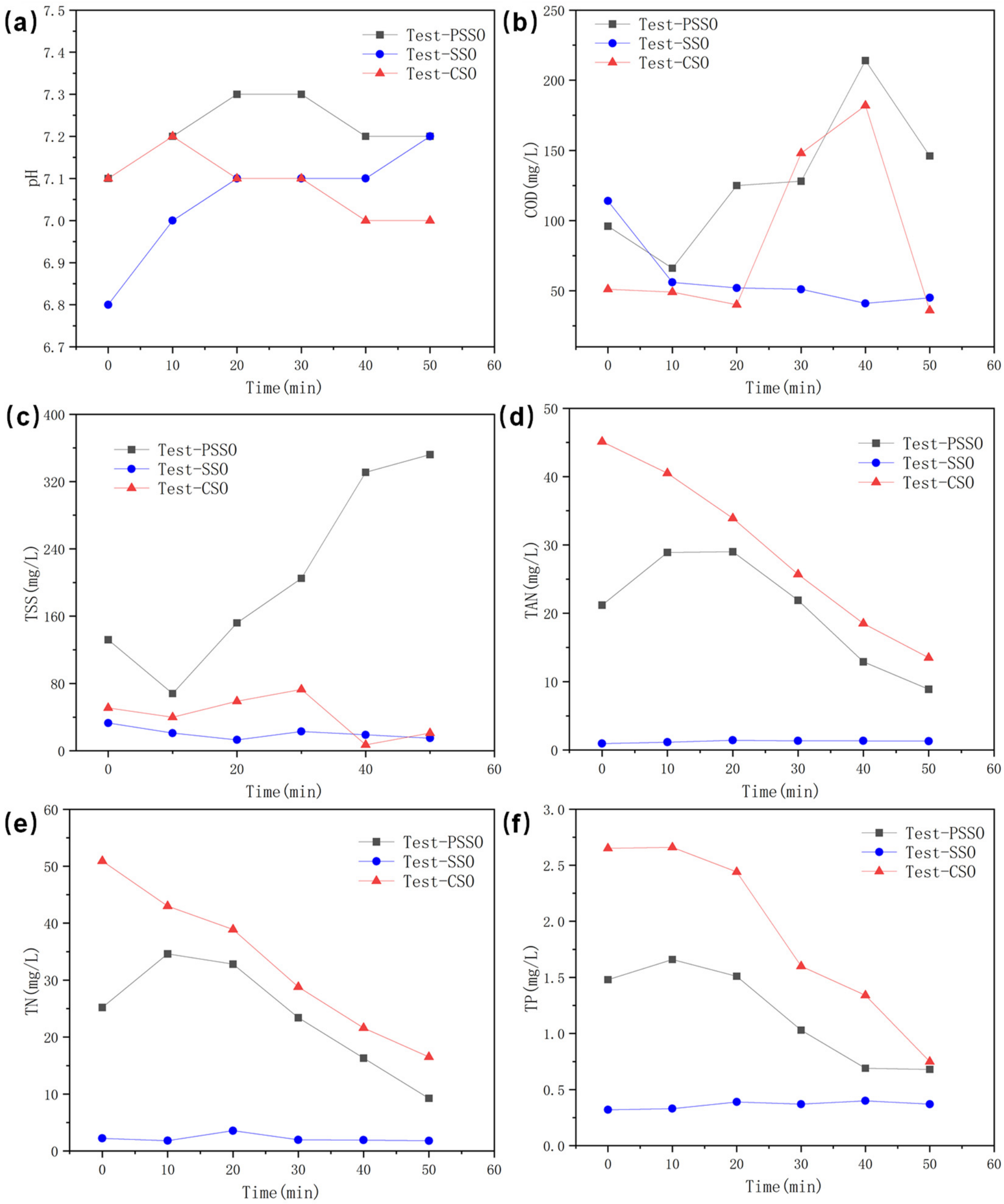

Figure 2. The water quality characteristics of the three target outlets in a short rainfall event: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

In summary, the main pollutants during overflow events across the three outlets followed the order: COD ≥ TSS > TN > TAN > TP. The separated sewer outlet experienced minimal overflow pollution, primarily sourced from rainwater. In contrast, rainwater worsened the overflow pollution at the combined sewer outlet by increasing COD and TSS but reducing nitrogen and phosphorus pollution intensity. The partially separated sewer outlet exhibited varying degrees of overflow pollution across all pollutants, mainly due to residual pipeline pollutants, with delayed but prolonged and more intense overflow pollution for COD and TSS.

3.1.2. Effect of the Partial Separation

A longer rainfall monitoring (110 min) was conducted at Test-PSSO and Test-SSO to analyze overflow pollution characteristics (Figure 3). At the Test-PSSO site, pH levels were consistently lower than at Test-SSO, but both remained weakly alkaline. The pH trend at Test-PSSO continuously rose, while it declined at Test-SSO as rainfall increased. This suggests that Test-PSSO contained more alkaline substances in its pipes, which interacted with rainwater during overflow.

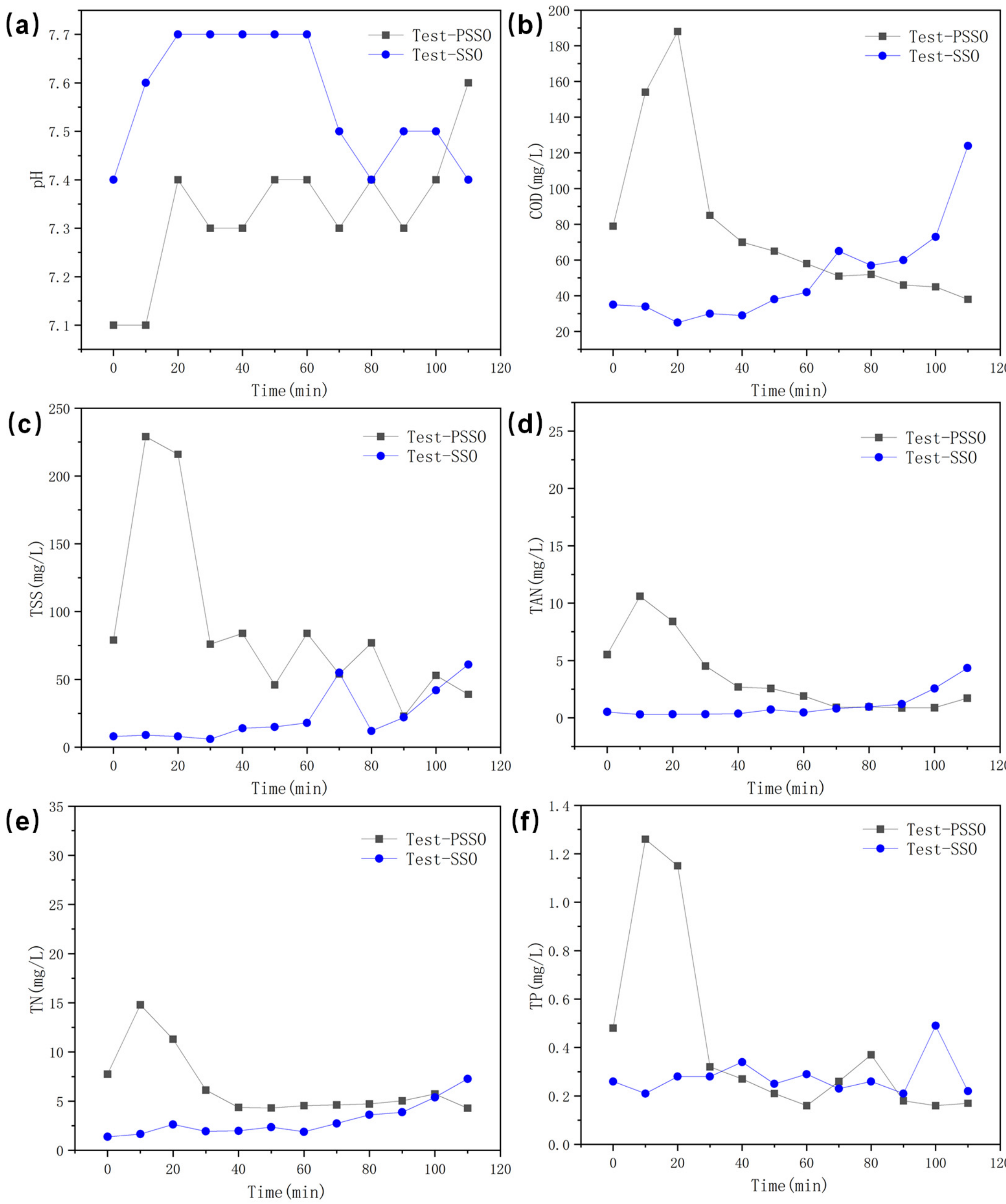

Figure 3. The water quality characteristics of Test-SSO and Test-PSSO in a long rainfall event: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

Pollutants such as TSS, TN, TP, COD, and TAN followed a similar pattern at each site. However, Test-PSSO showed a sharp increase in pollutant concentrations, which later declined to similar levels to that of Test-SSO, while Test-SSO exhibited a gradual increase. This suggests that the pollutants at Test-PSSO, which were retained in its pipes during dry periods, were flushed out by rainwater, but the rainwater will be the main source of pollution as the rainfall increases. In contrast, the slower increase at Test-SSO indicated that runoff from various underlying surfaces was the main contributor.

The variation in each pollutants' concentrations indicated that the Test-SSO achieved complete separation of rainwater and sewage, and the overflow pollution at this outlet is not obvious. However, the first flush overflow pollution at Test-PSSO occurred rapidly, peaking within 10 min, and stabilizing after 30 min, indicating that the initial rainfall caused significant overflow pollution that lasted for 30 min at least. The overflow pollution characteristics at Test-PSSO was similar to that observed during the shorter rainfall (50 min). However, this rainfall event presented an advanced peak time and shorter duration for COD and TSS, possibly due to the higher rainfall intensity [18,23]. The rainfall of this rainfall event was 20 mm, while that of the shorter rainfall event was 3.8 mm. The higher rainfalls led to the stronger scour of the pipeline pollutants and the higher overflow pollution intensity.

3.1.3. Effects of the Combined Sewer

Longer rainfall monitoring (230 min) was conducted at Test-PSSO and Test-CSO to analyze the impact of combined sewer systems on overflow pollution (Figure 4). The pH at Test-CSO was higher than at Test-PSSO, mainly influenced by the characteristics of the sewage from the nearby residential areas, as Test-CSO was located near a food market in the old urban area.

At Test-CSO, the concentrations of all pollutants were significantly higher than that at Test-PSSO, indicating the greater severity of combined sewer overflows. TAN and TN concentrations decreased over time, suggesting that the rainwater with lower nitrogen contents diluted these dissolved pollutants [19]. However, COD, TSS, and TP concentrations increased significantly with rainfall. This was closely related to the nature of sewage and rainwater. The inflow of rainwater rich in these pollutants worsened the overflow pollution, creating a dual pollution effect of sewage and rainwater. Importantly, the previous study reported that the concentrations of pollutants in the rainfall runoff of the market area was much higher than that of other urban underlying surfaces [24]. The sewer sediment was another important contributor to the overflow pollution. Some previous studies demonstrated that the sewer sediment was the main source of overflow pollutants [25–27], with a contribution of 30%–80% [19]. Among the various pollutants, the contribution rate of sewer sediment was 20.9%–44.6% for TN, 35.66%–47.3% for TP, 35%–66% for TSS, and 24%–65% for COD; however, the contributions differ under different rainfall intensities and scouring flow rates [19,28–32]. Therefore, the control of sewer sediment should also be given more attention in the overflow pollution from combined sewer.

The pollutant behavior at Test-CSO during the longer rainfall was similar to the shorter rainfall events, but the peak times of COD and TSS concentrations were advanced, and the duration of higher pollutant concentrations was longer, due to the increased rainfall (12.2 mm). As the causes of overflow pollution are different from Test-PSSO, the stronger the rainfall intensity is, the more rainwater with higher concentrations of COD and TSS entered the overflow of Test-CSO, the longer the overflow pollution time, and the faster the concentration increased.

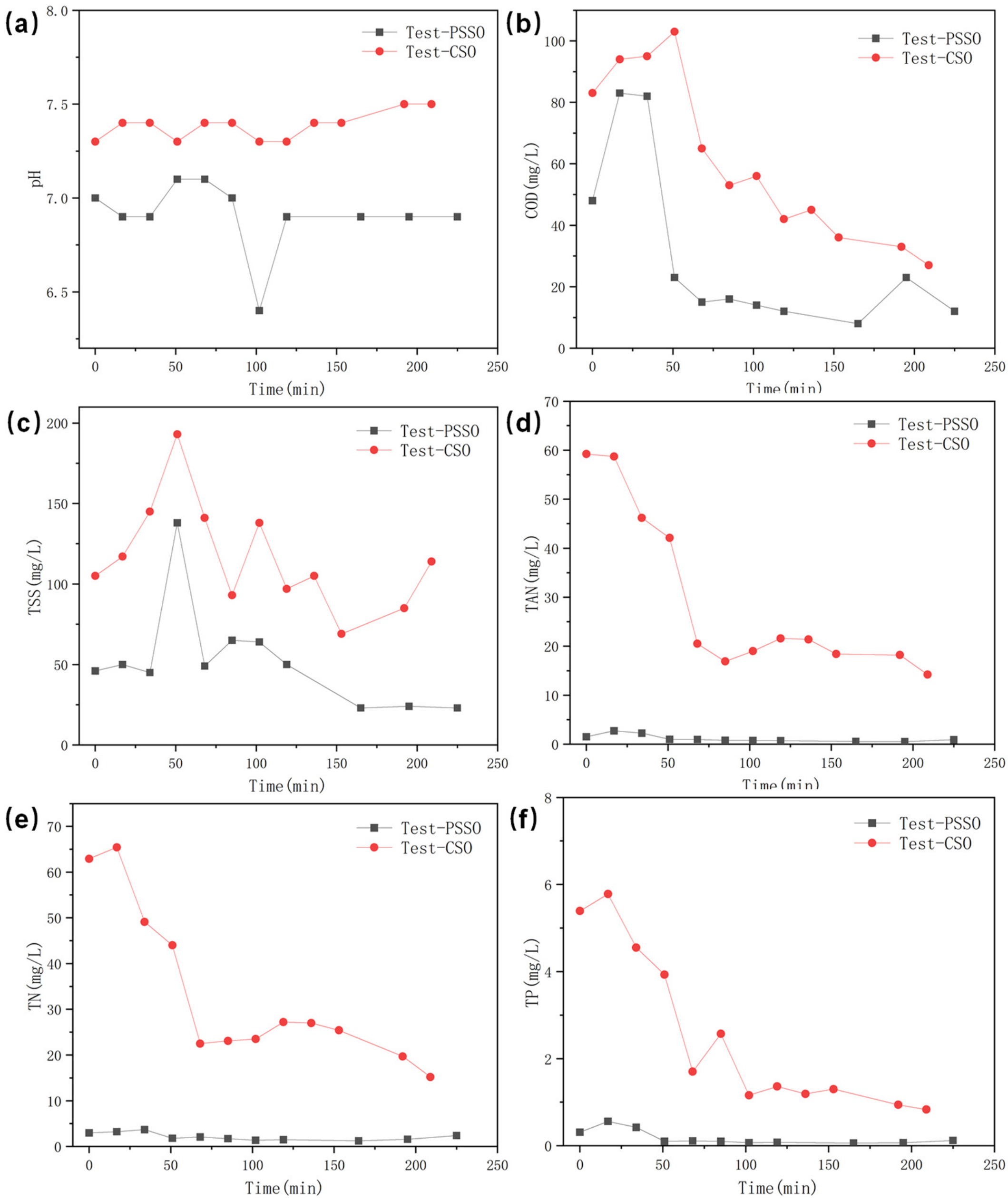

Figure 4. The water quality characteristics of Test-PSSO and Test-CSO in a long rainfall event: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

3.2. Analysis of Key Influencing Factors of Overflow Pollution

Rainfall is a crucial parameter affecting overflow pollution. The present study conducted the Pearson correlation analysis on the rainfall, the peak time, the overflow duration, and the peak concentration. Since the first flush overflow pollution was not significant at Test-SSO and the discharge of Test-CSO was all considered as overflow pollution, they were excluded from the analysis.

The analysis showed that rainfall was negatively correlated with the peak time and the overflow duration at Test-PSSO, particularly for COD and TSS (Table 1). This confirmed that higher rainfall led to earlier overflow peaks, which was consistent with the results in Section 3.1.2. However, the correlation between rainfall and overflow duration were weaker for most pollutants, despite that noted for TP. The correlation between rainfall and the peak concentration was very low. These were mainly because the overflow duration and peak concentration were greatly affected by the total amount of residual pollutants in the pipeline. The greater the total amount of residual pollutants is, the longer the overflow duration, and the higher the peak concentration correspondingly. In fact, the peak time, peak concentration, and duration were affected by many factors, such as spatial scales [33]. The larger the catchment area, the more random and complex the transmission and confluence process of runoff would be, further affecting the peak time and peak pollutant concentration [34–37].

Table 1. Pearson correlation between rainfall and key parameters.

Pearson Correlation	COD	TSS	TN	TAN	TP
Peak Time	−0.89	−0.90	−0.61	−0.59	−0.54
Duration	−0.02	−0.21	−0.48	−0.35	−0.72
Peak Con.	0.27	0.17	−0.17	−0.20	0.22

The dry period preceding each rain event was another important parameter affecting the pollution caused by the stormwater runoff [38]. The present study conducted the Pearson correlation analysis on the dry period, the peak time, the overflow duration, and the peak concentration (Table 2). The results showed that the dry period was positively correlated with the peak time but had a weakly negtive correlation with the peak concentration of pollutants. Some studies have demonstrated that the pollutant concentrations in the stormwater runoff increase with the extension of the dry period [39,40]. The weakly negtive correlation observed in the present study indicated that the pollutants in the overflow pollution of Test-PSSO were mainly from the pipe sediment, which was consistent with the results in Section 3.1.2. The dry period had a strongly positive influence on the duration of TN, TAN, and TP, but the correlation with COD and TSS was very low. These suggested that the dry period affected both the accumulation of TN, TAN, and TP in the piple sediment and the pollutant concentration in the stormwater runoff. However, the parameters affecting the characteristics of COD and TSS in the overflow pollution events were complex.

Table 2. Pearson correlation between dry period and the key parameters.

Pearson Correlation	COD	TSS	TN	TAN	TP
Peak time	0.70	0.81	0.86	0.68	0.91
Duration	−0.34	−0.13	0.80	0.63	0.90
Peak Con.	−0.46	−0.48	−0.26	−0.23	−0.50

3.3. Traceability Analysis of Overflow Pollution

The water quality of the rainwater was directly influenced by the functional characteristics of urban areas and the types of underlying surfaces. Therefore, the present study analyzed the runoff of the roof, grassland, and road from industrial parks, residential areas, and commercial areas to trace the sources of overflow pollution in a rainfall event (27.5 mm).

3.3.1. Effects of Different Underlying Surfaces in Industrial Parks

In industrial parks (Figure 5), COD concentrations were similar across different underlying surfaces. The pH in this area had the characteristics of acidification under different underlying surfaces, especially road runoff, but grassland had the ability to mitigate the pH decline. Nitrogen pollutants were highest on roofs, while grasslands showed the lowest nitrogen concentrations. The results showed that grassland had a certain ability to alleviate the nitrogen concentration in the rainwater, and the situation was also similar for total phosphorus. In terms of TP, although the highest concentrations were observed in the road runoff. The highest concentrations were less than 0.15 mg/L, indicating that the environmental impact was small. TSS levels were the highest in grassland runoff since there was no reasonable drainage channel in this grassland, and its runoff contained a large amount of grassland floating soil. When the floating soil was washed out, the concentrations of TSS decreased sharply. Overall, the main pollutants in the industrial park followed the order: TSS > COD > TN > TAN > TP. The main pollution underlying surfaces followed the order: roof > road > grass. Additionally, the concentrations of all pollutants decreased with the rainfall time.

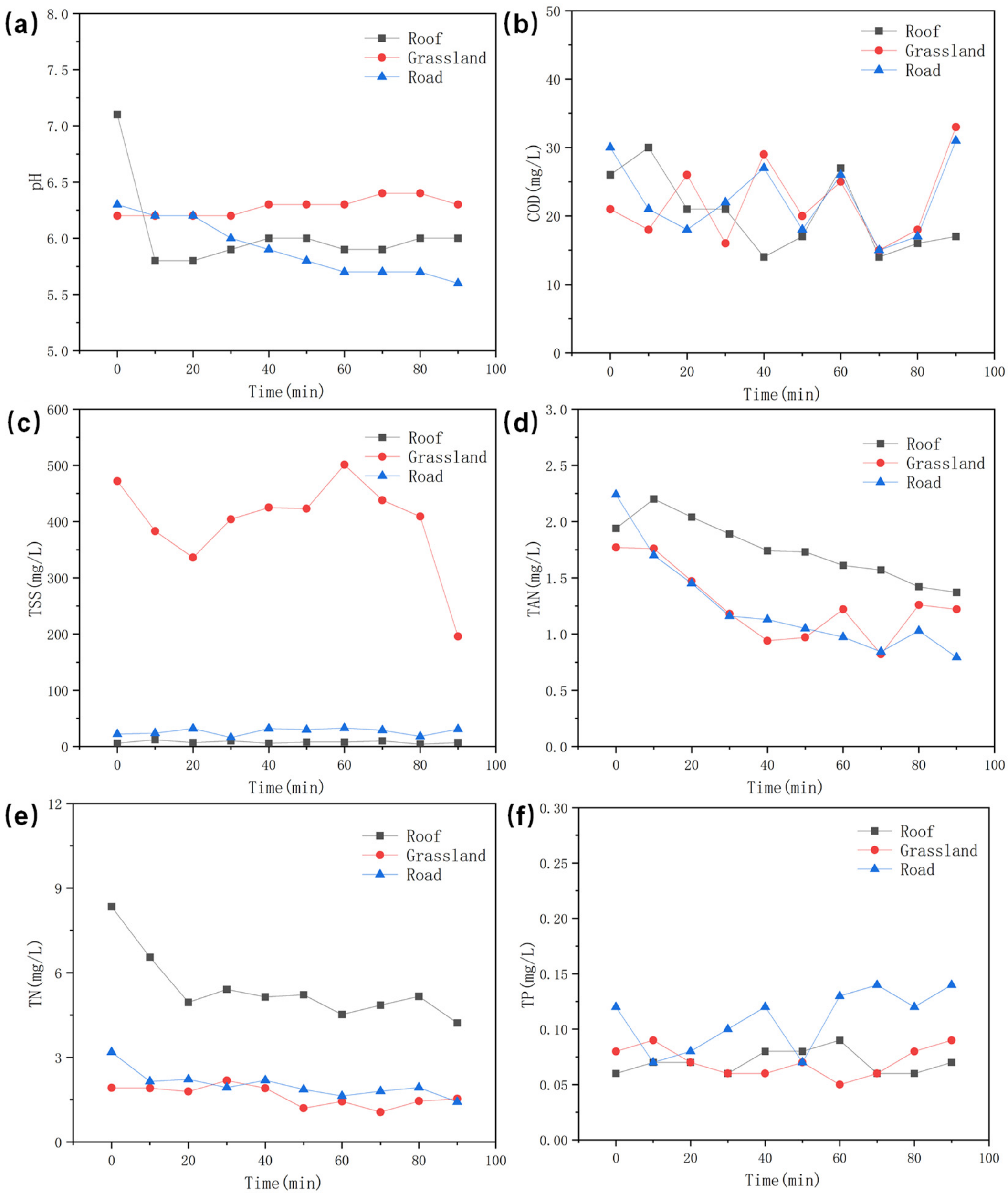

Figure 5. The water quality characteristics of different underlying surfaces in industrial parks: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

3.3.2. Effects of Different Underlying Surfaces in Residential Areas

In residential areas (Figure 6), the main pollution underlying surfaces followed the same order as industrial parks: roof > road > grassland. The concentrations of all pollutants decreased with the rainfall time, following the order: COD > TSS > TN > TAN > TP. The grassland had a better drainage system, and the concentrations of TSS decreased significantly compared with that in the industrial park. Therefore, the grassland with a good drainage system had an effective buffering effect on rainwater pollution. Moreover, the infiltration capacity of grassland was significantly affected by the maintenance practices. The study by Galli et al. [41] declared that the unsaturated hydraulic conductivity may decrease rapidly after about 9–12 years in the absence of soil and vegetation maintenance. Soil rehabilitation and vegetation cover were efficient methods to improve the infiltration capacity of grassland. The grassland in the residential area had better and more frequent maintenance by the specialized management organizations than that in the industrial park, resulting in the higher infiltration capacity. However, the concentrations of various pollutants on the other underlying surfaces in the residential area were higher than that in the industrial park, indicating that the industrial park had effective pollution control measures and its impact on the environment was significantly reduced. In contrast, the residential area had more frequent human activities, and its impact on the environment was more significant.

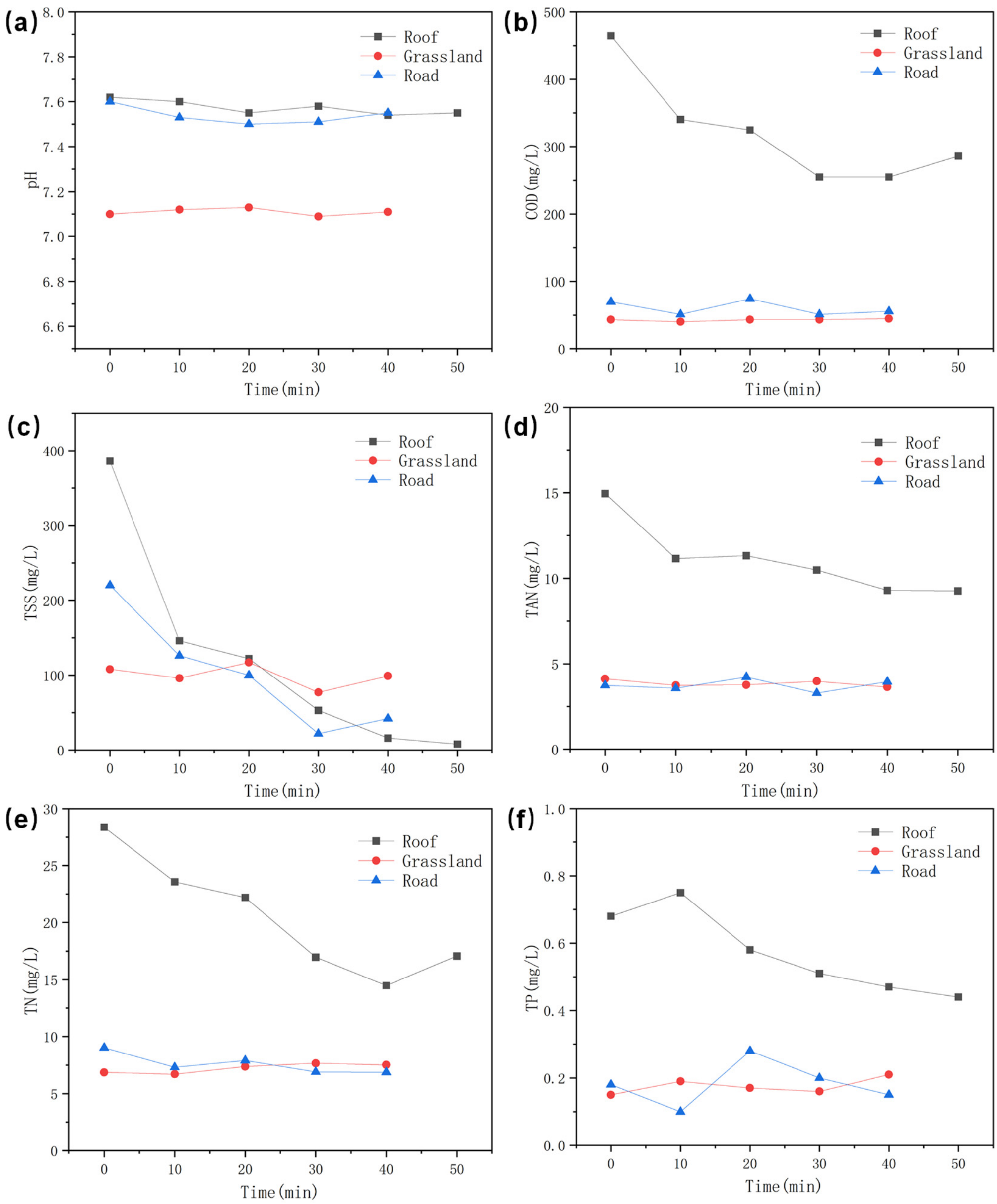

Figure 6. The water quality characteristics of different underlying surfaces in residential areas: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

3.3.3. Effects of Different Underlying Surfaces in Commercial Areas

In commercial areas (Figure 7), the main pollution underlying surfaces for nitrogen pollutants followed the order: roof > road > grassland. Among them, the TAN was the main nitrogen form. In terms of COD, TSS, and TP, the pollutant concentrations in the road runoff was the highest, while the roof contributed the least, which was mainly due to the large levels of human traffic on the road in the commercial area. Grasslands buffered pH and pollutant concentrations, reducing the environmental impact. The main pollutants in the commercial areas followed the order: TSS > COD > TN > TAN > TP.

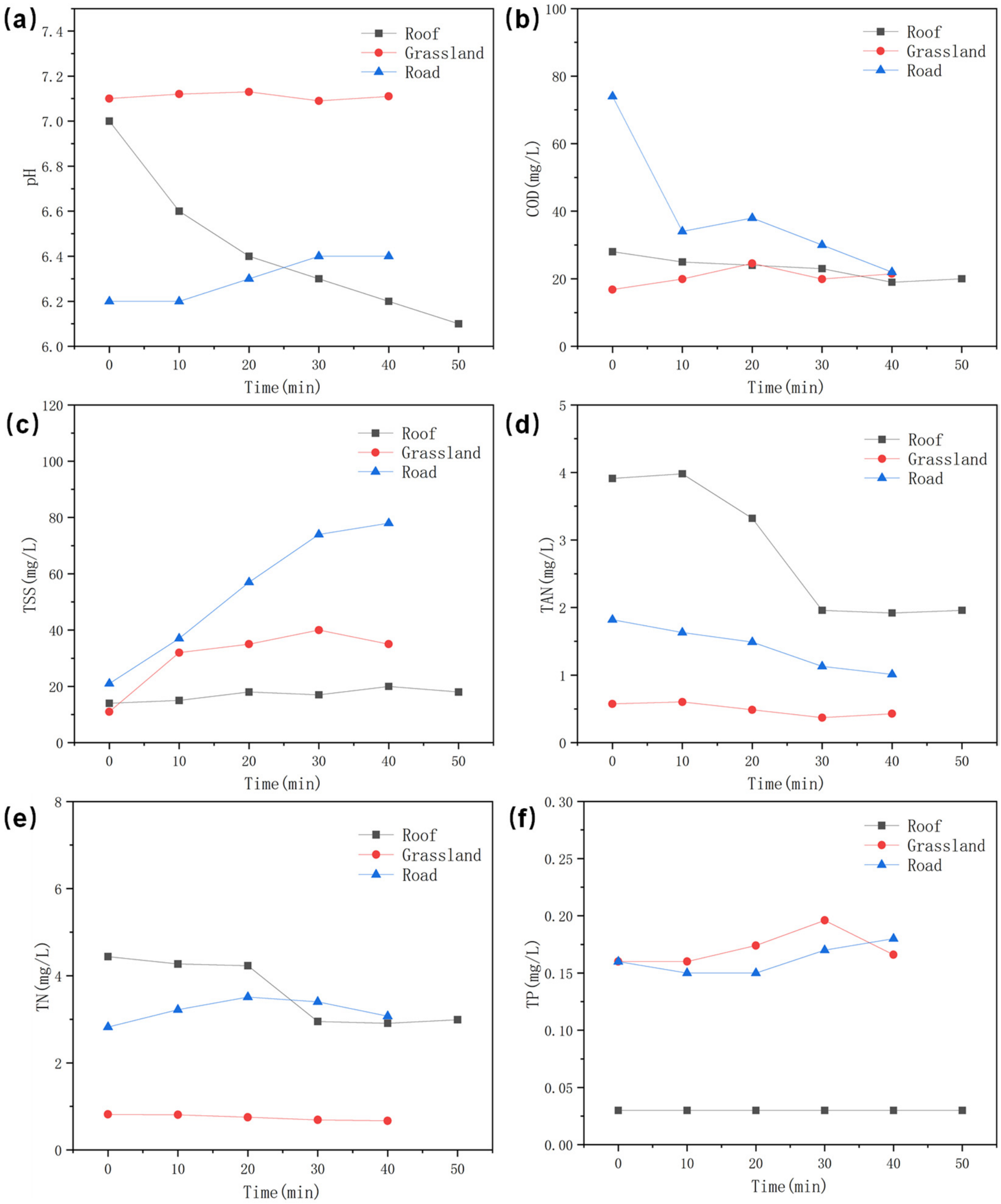

Figure 7. The water quality characteristics of different underlying surfaces in commercial areas: (**a**) pH; (**b**) COD; (**c**) TSS; (**d**) TAN; (**e**) TN; (**f**) TP.

Across the three functional areas, the pollution intensity exhibited the following order: residential areas > industrial parks > commercial areas. Among them, the residential areas were significantly higher than the other two areas in terms of various pollutants. With the exception of commercial areas, the pollution intensity of underlying surfaces generally followed the order: roof > road > grassland. The main pollutants were COD and TSS. TAN was the dominant nitrogen form, representing was over 50% of TN, while TP contributed less to the total pollution intensity with very low concentrations. These results were consistent with the study by Lai et al. [24], in which the concentrations of COD and TSS noted on the roof were higher than that of other underlying surfaces in an urban residential area. The pollutant concentrations on the different underlying surfaces in each functional area decreased gradually with the rainfall time, indicating that the pollution intensity of the initial rain was the highest. The first flush effect has also been observed in some studies, in which the initial runoff is disproportionately carrying the most of the pollutants in the whole runoff, resulting in the fact that the concentration of pollutants in the early stage of rainfall runoff is significantly higher than that in the later stage [42–45]. However, the first flush effect would decrease with the increasing of rainfall intensity and is even not obvious under moderate and heavy rain conditions, especially on the underlying surfaces of roof and road [24,46]. Notably, grasslands provided a buffering effect, reducing pollutant concentrations and pH fluctuations, highlighting the importance of green spaces in controlling rainwater pollution, which was consistent with the study by Wang et al. [46]. The above results were also consistent with the description of the rainwater in Section 3.1, and the variation trend of the concentrations of pollutants in the rainwater, especially roof runoff (gradually decreasing to stability), was similar to the trend of pollutants at rainwater outlets (gradually decreasing to stability), indicating that the runoff from the roof was an important contributor to the overflow pollution. In addition, the TSS and COD in the rainwater contributed the most to the overflow pollution intensity.

4. Conclusions

The present study explored the characteristics and sources of overflow pollution during the flood season by monitoring three types of rainwater outlets: combined sewer outlet, separated sewer outlet, and partially separated sewer outlet. The results revealed that the primary pollutants during overflow events across all outlets were COD and TSS. The separated sewer outlet experienced minimal overflow pollution, while rainwater exacerbated the overflow pollution at the combined sewer outlet by increasing concentrations of COD and TSS but reducing the pollution intensity of nitrogen and phosphorus. The partially separated sewer outlet showed varying degrees of overflow pollution for all types of pollutants, primarily due to residual contaminants in the pipelines. The peak time was negatively correlated with the rainfall, meaning higher rainfall led to quicker peaks at partially separated sewer outlets.

Traceability analysis showed that residential areas had significantly higher pollutant concentrations on various underlying surfaces compared to the industrial park and the commercial area. Despite that noted for the commercial area, the pollution intensity of underlying surfaces followed the order of roof > road > grassland, with COD and TSS being the main pollutants, followed by nitrogen compounds, where TAN was the primary form of nitrogen. The runoff from roofs was identified as a significant contributor to the overflow pollution of rainwater outlets. Grasslands were found to provide an important buffering effect, reducing the concentration of nitrogen, phosphorus, and COD, while also stabilizing pH levels, which underscored the role of green spaces in mitigating rainwater pollution during initial rain events.

The results emphasized the need for targeted control measures to reduce the impact of overflow pollution during the flood season. Effective separation of rainwater and sewage systems, increased green space to buffer pollutants, and better management of roof runoff could significantly reduce pollution intensity and improve urban water quality.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w16223159/s1. Table S1: The water quality of the three outlets and rainfall information during the monitoring period.

Author Contributions: Conceptualization, S.Y., H.X., Y.F. and J.L.; methodology, Y.F.; software, Y.F.; validation, Y.F., M.L., G.L., L.H., Y.L. and G.S.; formal analysis, Y.F.; investigation, J.L., M.L., G.L., L.H., Y.L. and G.S.; resources, S.Y., H.X. and Y.F.; data curation, Y.F.; writing—original draft preparation, S.Y. and H.X.; writing—review and editing, S.Y. and H.X.; visualization, Y.F.; supervision, S.Y. and H.X.; project administration, S.Y. and H.X.; funding acquisition, S.Y., H.X. and Y.F. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Henan Provincial Technical Center for Ecology and Environment Research Project (No. 2023), Key Research and Development Project of Henan Province (No. 221111320900), Science and Technology Research Project of Henan Province (No. 222102320389), Graduate Education Reform and Quality Improvement Project of Henan Province (No. YJS2023AL012), Science and Technology Project of Henan Provincial Department of Transportation (No. 2023-4-4), Scientific Research Foundation for Academician of CAS Team of Zhengzhou University (No. 13432340370), and research funding from Zhengzhou University (No. 32213813).

Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

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

References

1. Ekhlas, D.; Kurisu, F.; Kasuga, I.; Cernava, T.; Berg, G.; Liu, M.; Furumai, H. Identification of new eligible indicator organisms for combined sewer overflow via 16S rRNA gene amplicon sequencing in Kanda River, Tokyo. *J. Environ. Manag.* **2021**, *284*, 112059. [CrossRef]
2. Joshi, P.; Leitao, J.P.; Maurer, M.; Bach, P.M. Not all SuDS are created equal: Impact of different approaches on combined sewer overflows. *Water Res.* **2021**, *191*, 116780. [CrossRef]
3. Madoux-Humery, A.S.; Dorner, S.M.; Sauvé, S.; Aboulfadl, K.; Galarneau, M.; Servais, P.; Prevost, M. Temporal analysis of E.coli, TSS and wastewater micropollutant loads from combined sewer overflows: Implications for management. *Environ. Sci.-Process. Impacts* **2015**, *17*, 965–974. [CrossRef]
4. Riechel, M.; Matzinger, A.; Pawlowsky-Reusing, E.; Sonnenberg, H.; Uldack, M.; Heinzmann, B.; Caradot, N.; von Seggern, D.; Rouault, P. Impacts of combined sewer overflows on a large urban river—Understanding the effect of different management strategies. *Water Res.* **2016**, *105*, 264–273. [CrossRef]
5. Venditto, T.; Ponzelli, M.; Sarathy, S.; Ray, A.K.; Santoro, D. A microsieve-based filtration process for combined sewer overflow treatment with nutrient control: Modeling and experimental studies. *Water Res.* **2020**, *170*, 115328. [CrossRef]
6. Botturi, A.; Daneshgar, S.; Cordioli, A.; Foglia, A.; Eusebi, A.L.; Fatone, F. An innovative compact system for advanced treatment of combined sewer overflows (CSOs) discharged into large lakes: Pilot-scale validation. *J. Environ. Manag.* **2020**, *256*, 109937. [CrossRef]
7. Phillips, P.J.; Chalmers, A.T.; Gray, J.L.; Kolpin, D.W.; Foreman, W.T.; Wall, G.R. Combined Sewer Overflows: An Environmental Source of Hormones and Wastewater Micropollutants. *Environ. Sci. Technol.* **2012**, *46*, 5336–5343. [CrossRef]
8. Viviano, G.; Valsecchi, S.; Polesello, S.; Capodaglio, A.; Tartari, G.; Salerno, F. Combined Use of Caffeine and Turbidity to Evaluate the Impact of CSOs on River Water Quality. *Water Air Soil Pollut.* **2017**, *228*, 330. [CrossRef]
9. Madoux-Humery, A.-S.; Dorner, S.; Sauve, S.; Aboulfadl, K.; Galarneau, M.; Servais, P.; Prevost, M. Temporal variability of combined sewer overflow contaminants: Evaluation of wastewater micropollutants as tracers of fecal contamination. *Water Res.* **2013**, *47*, 4370–4382. [CrossRef]
10. Liao, Z.-L.; Zhao, Z.-C.; Zhu, J.-C.; Chen, H.; Meng, D.-Z. Complexing characteristics between Cu(II) ions and dissolved organic matter in combined sewer overflows: Implications for the removal of heavy metals by enhanced coagulation. *Chemosphere* **2021**, *265*, 129023. [CrossRef]
11. Arshad, M.; Silvestre, J.; Pinelli, E.; Kallerhoff, J.; Kaemmerer, M.; Tarigo, A.; Shahid, A.; Guiresse, M.; Pradere, P.; Dumat, C. A field study of lead phytoextraction by various scented *Pelargonium* cultivars. *Chemosphere* **2008**, *71*, 2187–2192. [CrossRef]
12. Gromaire, M.C.; Garnaud, S.; Saad, M.; Chebbo, G. Contribution of different sources to the pollution of wet weather flows in combined sewers. *Water Res.* **2001**, *35*, 521–533. [CrossRef]
13. Gosset, A.; Ferro, Y.; Durrieu, C. Methods for evaluating the pollution impact of urban wet weather discharges on biocenosis: A review. *Water Res.* **2016**, *89*, 330–354. [CrossRef]
14. Taebi, A.; Droste, R.L. Pollution loads in urban runoff and sanitary wastewater. *Sci. Total Environ.* **2004**, *327*, 175–184. [CrossRef]
15. Alyaseri, I.; Zhou, J.; Morgan, S. Sustainable stormwater management using rain gardens in urban areas. IOP Conf. Ser., Earth Environ. *Sciecne* **2021**, *779*, 012041. [CrossRef]
16. Gao, Y.; Shi, X.; Jin, X.; Wang, X.C.; Jin, P. A critical review of wastewater quality variation and in-sewer processes during conveyance in sewer systems. *Water Res.* **2023**, *228*, 119398. [CrossRef]
17. APHA. *Standard Methods for the Examination of Water and Wastewater*, 21st ed.; American Public Health Association: Washington, DC, USA, 2005.

18. Li, Y.; Zhou, Y.; Wang, H.; Jiang, H.; Yue, Z.; Zheng, K.; Wu, B.; Banahene, P. Characterization and sources apportionment of overflow pollution in urban separate stormwater systems inappropriately connected with sewage. *J. Environ. Manag.* **2022**, *303*, 114231. [CrossRef]
19. Chen, Y.; Shi, X.; Jin, X.; Jin, P. Characteristics of overflow pollution from combined sewer sediment: Formation, contribution and regulation. *Chemosphere* **2022**, *298*, 134254. [CrossRef]
20. Niazkar, M.; Evangelisti, M.; Peruzzi, C.; Galli, A.; Maglionico, M.; Masseroni, D. Investigating First Flush Occurrence in Agro-Urban Environments in Northern Italy. *Water* **2024**, *16*, 891. [CrossRef]
21. Shi, X.; Sang, L.T.; Wang, X.C.C.; Jin, P.K. Pollutant exchange between sewage and sediment in urban sewer systems. *Chem. Eng. J.* **2018**, *351*, 240–247. [CrossRef]
22. Bertrand-Krajewski, J.L.; Chebbo, G.; Saget, A. Distribution of pollutant mass vs volume in stormwater discharges and the first flush phenomenon. *Water Res.* **1998**, *32*, 2341–2356. [CrossRef]
23. Yin, H.; Lu, Y.; Xu, Z.; Li, H.; Schwegler, B.R. Characteristics of the overflow pollution of storm drains with inappropriate sewage entry. *Environ. Sci. Pollut. Res.* **2016**, *24*, 4902–4915. [CrossRef]
24. Lai, Q.Y.; Ma, J.; Du, W.; Luo, Y.D.; Ji, D.W.; He, F. Analysis of the Source Tracing and Pollution Characteristics of Rainfall Runoff in Adjacent New and Old Urban Areas. *Water* **2023**, *15*, 3018. [CrossRef]
25. Kaeseberg, T.; Schubert, S.; Oertel, R.; Zhang, J.; Berendonk, T.U.; Krebs, P. Hot spots of antibiotic tolerant and resistant bacterial subpopulations in natural freshwater biofilm communities due to inevitable urban drainage system overflows. *Environ. Pollut.* **2018**, *242*, 164–170. [CrossRef]
26. Meng, D.; Jin, W.; Chen, K.; Zhang, C.; Zhu, Y.; Li, H. Cohesive strength changes of sewer sediments during and after ultrasonic treatment: The significance of bound extracellular polymeric substance and microbial community. *Sci. Total Environ.* **2020**, *723*, 138029. [CrossRef]
27. Xu, Z.; Wu, J.; Li, H.; Chen, Y.; Xu, J.; Xiong, L.; Zhang, J. Characterizing heavy metals in combined sewer overflows and its influence on microbial diversity. *Sci. Total Environ.* **2018**, *625*, 1272–1282. [CrossRef]
28. Al Aukidy, M.; Verlicchi, P. Contributions of combined sewer overflows and treated effluents to the bacterial load released into a coastal area. *Sci. Total Environ.* **2017**, *607*, 483–496. [CrossRef]
29. Crocetti, P.; Eusebi, A.L.; Bruni, C.; Marinelli, E.; Darvini, G.; Carini, C.B.; Bollettini, C.; Recanati, V.; Akyol, C.; Fatone, F. Catchment-wide validated assessment of combined sewer overflows (CSOs) in a mediterranean coastal area and possible disinfection methods to mitigate microbial contamination. *Environ. Res.* **2021**, *196*, 110367. [CrossRef]
30. David, T.; Borchardt, D.; von Tuempling, W.; Krebs, P. Combined sewer overflows, sediment accumulation and element patterns of river bed sediments: A quantitative study based on mixing models of composite fingerprints. *Environ. Earth Sci.* **2013**, *69*, 479–489. [CrossRef]
31. Hata, A.; Katayama, H.; Kojima, K.; Sano, S.; Kasuga, I.; Kitajima, M.; Furumai, H. Effects of rainfall events on the occurrence and detection efficiency of viruses in river water impacted by combined sewer overflows. *Sci. Total Environ.* **2014**, *468*, 757–763. [CrossRef]
32. Liu, C.Y.; Yang, Y.T.; Zhou, J.Q.; Chen, Y.Z.; Zhou, J.; Wang, Y.Y.; Fu, D.F. Migration and transformation of nitrogen in sediment-water system within storm sewers. *J. Environ. Manag.* **2021**, *287*, 112355. [CrossRef]
33. Wang, J.; Zhao, L.; Zhang, X. First flush effect analysis of urban storm runoff in combined sewer system. *Environ. Pollut. Control* **2015**, *37*, 12–20.
34. Hai, Y.; Dian, L.; Yu, D.; Wei, Y.; Liu, M. Effect of rainfall on the pollution characteristics of combined sewer overflows. *Chin. J. Environ. Eng.* **2020**, *14*, 3082–3091.
35. Li, H.; Xu, S.; Huang, Y.; Wei, P. Pollution loading of overflow in combined drainage channels during rainy season. *Acta Sci. Circumstantiae* **2013**, *33*, 2522–2530.
36. Xu, Z.X.; Xiong, L.J.; Li, H.Z.; Liao, Z.L.; Yin, H.L.; Wu, J.; Xu, J.; Chen, H. Influences of rainfall variables and antecedent discharge on urban effluent concentrations and loads in wet weather. *Water Sci. Technol.* **2017**, *75*, 1584–1598. [CrossRef]
37. Xu, Z.X.; Xiong, L.J.; Li, H.Z.; Yin, H.L.; Wu, J.; Xu, J.; Zhang, J. Pollution characterization and source analysis of the wet weather discharges in storm drainages. *Desalin. Water Treat.* **2017**, *72*, 169–181. [CrossRef]
38. Kim, L.H.; Kayhanian, M.; Zoh, K.D.; Stenstrom, M.K. Modeling of highway stormwater runoff. *Sci. Total Environ.* **2005**, *348*, 1–18. [CrossRef]
39. Vermette, S.J.; Irvine, K.N.; Drake, J.J. Temporal variability of the elemental composition in urban street dust. *Environ. Monit. Assess.* **1991**, *18*, 69–77. [CrossRef]
40. Yuan, Y.; Hall, K.; Oldham, C. A preliminary model for predicting heavy metal contaminant loading from an urban catchment. *Sci. Total Environ.* **2001**, *266*, 299–307. [CrossRef]
41. Galli, A.; Peruzzi, C.; Beltrame, L.; Cislaghi, A.; Masseroni, D. Evaluating the infiltration capacity of degraded vs. rehabilitated urban greenspaces: Lessons learnt from a real-world Italian case study. *Sci. Total Environ.* **2021**, *787*, 147612. [CrossRef]
42. Hathaway, J.M.; Tucker, R.S.; Spooner, J.M.; Hunt, W.F. A Traditional Analysis of the First Flush Effect for Nutrients in Stormwater Runoff from Two Small Urban Catchments. *Water Air Soil Pollut.* **2012**, *223*, 5903–5915. [CrossRef]
43. Ma, Z.B.; Ni, H.G.; Zeng, H.; Wei, J.B. Function formula for first flush analysis in mixed watersheds: A comparison of power and polynomial methods. *J. Hydrol.* **2011**, *402*, 333–339. [CrossRef]

44. Ou, L.-B.; Hu, D.; Huang, Y.; Cui, S.-Y.; Guo, T.-J.; Zhang, W.; Wang, X.-J. First flush analysis of PAHs in roof runoff in Beijing. *Huan Jing Ke Xue* **2011**, *32*, 2896–2903. [PubMed]
45. YuFen, R.E.N.; XiaoKe, W.; Bing, H.A.N.; ZhiYun, O.; Hong, M. Chemical analysis on stormwater-runoffpollution of different underlying urban surfaces. *Acta Ecol. Sin.* **2005**, *25*, 3225–3230.
46. Wang, Z.; Lei, G.Y.; Niu, Y. Iop, The first flush effect of different urban underlying surfaces through artificial simulated rainfall. In Proceedings of the International Symposium on Resource Exploration and Environmental Science (REES), Ordos, China, 14–16 April 2017; p. 012077.