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To cite this article: Chunyi Duan and Hong Chen 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **252** 052080

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Research on Arsenic Release in Ground-Water-Based Distribution System

Chunyi Duan *, Hong Chen

Guangxi Polytechnic of Construction, 33 Luowen Road, Nanning, 530000, China

*Corresponding author e-mail: dcyjoyce@126.com

Abstract. The concentration of arsenic in effluent can meet standard requirements for drinking water after strengthening arsenic removal process, which is less than $10 \mu\text{g/L}$ and stability in $5 \sim 8 \mu\text{g/L}$. However, residual arsenic deliveries and releases in water distribution system by particle adsorption, migration, desorption and solution effect. Resulting in the water users' arsenic concentration increases. This research aims to study the arsenic enrichment characteristics, arsenic release rule in water distribution system and the relationship between arsenic release and the other water quality parameters through the network residual arsenic census and monitoring indexes of water quality.

1. Introduction

Arsenic in drinking water is now a critical issue for the world. Arsenic has been ranked as the most formidable pollutant throughout the world. In the last few decades, the chemicals for As removal is Fe salt general. The effectiveness of Fe salt for As removal is strong adsorption capacity of Fe particle surface to As. There is a variety of iron particles in the water distribution system, such as pipeline corrosion byproduct. With the growth of the running time of the pipe network, iron particles will adsorb arsenic, and arsenic enrichment occurs in the wall. These particles release again to the water distribution system due to the hydraulic or chemical condition change, again arsenic release to the water pipe network, which will lead the arsenic concentration increased. The released arsenic may be particulate or solubility, the particles of arsenic release means interference and transmission of the particulate matter in the water containing arsenic. There are few studies on the elevated concentration of arsenic in the water supply network.

The first reported about arsenic release in pipe networks is in Nebraska, in the United States in 1997, from the chlorination triggered to the chlorine-free groundwater, the consequences of the outbreak of the entire pipe network "yellow water" accident. Accident during water sampling survey results showed that the iron pipe network released the dramatic elevated arsenic concentration to cause accidents mainly due to the "yellow water". The increase releasing of concentration of arsenic particles caused by dissolution is directly related. The arsenic release occurs in arsenic accident segment. Particles collected in the Midwestern United States 67 measurable arsenic pipe network observational studies, reported in the pipe network elements composition of the particles, which arsenic concentration from 10 to 13650 ($\mu\text{g As}/(\text{g solids})$) range. Studies have shown that, the particles collected in almost all cases, the content of Fe is the most abundant. Assumptions As is specific association under certain conditions by the constraints of Fe. Fe particles and sediment in the pipe network is usually from the water purification



plant residues or raw water or iron pipeline corrosion byproducts. Waterworks usually detect pH, phosphate as a corrosion inhibitor for evaluating changes in water quality. Furthermore, waterworks has a means of removal of arsenic. These factors could alter the balance of arsenic release conditions and pipe network arsenic mobility and fundamentally determines the user leading water arsenic levels. On the base of Abhijit which had conducted a detailed study on 9 pipe networks in the cycle 168h in the Midwest of U.S. The results show that the arsenic release in pipe network is solid surface desorption rather than dissolution of As. Arsenic release as the initial concentration of arsenic growth in the solid and the pH (tested range of 7 to 9) rises. pH will change the surface charge of Fe, occurred in the reverse reaction, which makes a rise of the water OH-1 concentration in water. When the pH of filtered water elevated, the surface of the iron solid adsorption potential change, the activity decreased and the desorption is enhanced. Meanwhile, in the various pH values under the conditions of the study, the presence of the presence of added as a corrosion inhibitor will increase the arsenic release. In addition, some non-chemical mechanisms of the process, such as the role of microorganisms, also have a certain influence on migrating As the pipe network.

In the microenvironment of pipe wall, arsenic element exists with the delicate chemical balance. The microenvironment of pipe wall includes pipes, pipe scale (arsenic compound) and the water environment. The chemical reaction process in the microenvironment of pipe wall comprises the arsenic release reaction and the deposition reaction of arsenic. As mentioned earlier, arsenic not only can be into the pipe network directly, but also be precipitated by the redox complex reaction in the tube wall to form a pipe scale. Pipe scale erosion in the pipe network hydraulic pipe network water release arsenic dissolution and other reasons, this is the arsenic release phenomenon. Arsenic in water network is the major release of corrosion products from the pipe wall.

The main significance lies in the study of this subject: Using laboratory established within the small pipe network to simulate the actual pipe network and sampling and water quality analysis, so that we can change indicators of water quality in the pipe network artificially, sampling conveniently and it's not affect the normal operation of actual water distribution system. Data analysis, laboratory pipe network data combined with the actual pipe network data analysis. The results of laboratory tests validated through actual pipe network data to achieve the purpose of guiding practice. Based on the pipe wall the quiz analysis of the growth of, certainly is enriched in a certain content of arsenic in the growth rings, the serious part of the pipeline and position enrichment. It is considered on this basis the erosion arsenic release test, aimed at the analysis pipeline network residual arsenic release law in a different water condition.

2. Material and methods

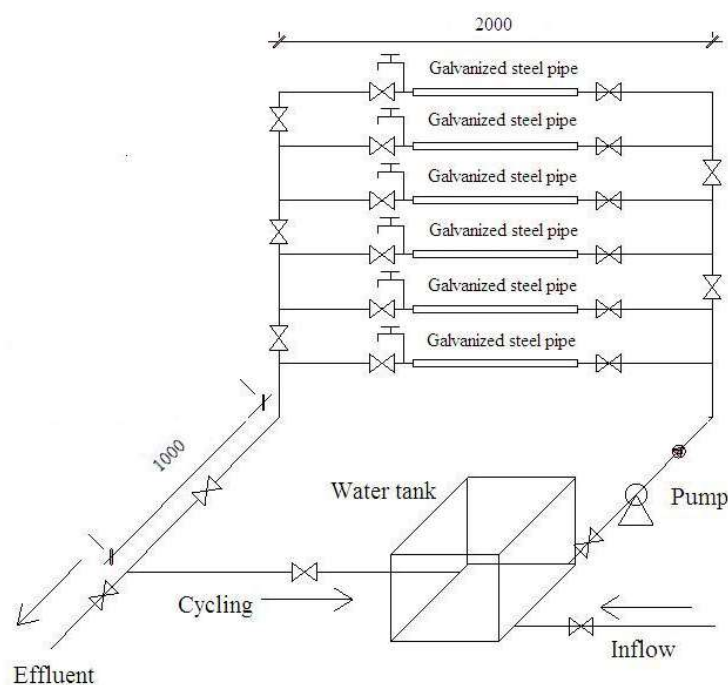
In the long term of transmission, irregular growth rings will form along the pipe wall gradually. It is a hybrid of sediment, rust material, sticky dirt and biofilm mutual combination. Growth rings are initial lose at first and can be washed away by the water flow easily, which cause the water outlet from users' to become the yellow water. Those which are not carried away accumulated and harden gradually to become a place of growth and reproduction of bacteria. Therefore, the component of the growth rings within the pipeline is likely to become a place of adsorption and enrichment of arsenic.

On 5 to July 2011, interception DN50 ~ DN500 various pipes and pipe wall growth ring component in the water supply area of the Eastern Zhou Dynasty waterworks for analysis and study the arsenic enrichment characteristics and release law in the water distribution.

Test devices simulated actual water distribution network construction, is shown in Figure 1. Each horizontal pipe equipped with a removable the biofilm samplers and a faucet for sampling. There are two valves mounted at the closed sampler of both ends of the pipe for closing easily when sampling from the cross pipe. 3 valves installed on each branch pipe riser, by switching valves we can achieve mutual switch between analog a small pipe network segment management.

Table 1. Instrument and analysis method

| Parameters | Analytical method | Instrumentation |
|-------------------------------|------------------------------|---|
| Water Temperature | Direct reading | Mercurial thermometer |
| Flow rate | Direct reading | rotameter |
| Turbidity | Turbidity meter | HACH-2100P Photoelectric turbidity meter |
| pH | Composite electrodes | SARTORUIS AG-PP-15E |
| Cl ⁻ | Chromatography of ions | DX-600 IC(ion chromatography) |
| SO ₄ ²⁻ | Id. | Id. |
| NO ₃ ⁻ | Id. | Id. |
| HPO ₄ ⁻ | Id. | Id. |
| F ⁻ | Id. | Id. |
| Fe | atomic emission spectrometry | Perkinelmer 5300DV Atomic emission spectrometer |
| Mn | Id. | Id. |
| Cr | Id. | Id. |
| Cu | Id. | Id. |
| Ca | Id. | Id. |
| Pb | Id. | Id. |
| Mg | Id. | Id. |
| As | Atomic fluorescence | JITIAN AFS 8220 Atomic fluorescence spectrophotometer |



Other pipelines are all PVC pipes in addition to label galvanized steel pipes which are labeled.

Figure 1. Washing - arsenic release reactor device

During the test, all the valves of the vertical pipes need to be opened so that the raw water in the tank can be transmitted to the other end of the horizontal pipes by the centrifugal pump, from the same direction of flow of the branch pipe to the other end of the respective transverse pipe, in a summary of the main pipe flow back into the tank as the cycle run. The characteristics of the parallel pipe network in running are water flow direction is transverse flow, the water effluent from the tank as the flow enters the entire branch pipes. The water in branch pipes is in the same flow direction and the hydraulic conditions of each is not the same (including the flow rate, and full flow or not) The time of raw water cycle which is through the path is relatively short and the flow rate of the pump is greater, pressure in the pipe network is lower. The operation cycle of the test simulated pipe network is two days around the clock. The water is changed 150L once for a cycle. Two main factors were considered in the test: the flow rate and pH. Velocity of flow increments from 0.3m/s to 1.3m/s during the flow test and each velocity period is running for 2h and sampling timing. The pH is changing between 7 to 9. Take the initial arsenic ions in raw water of the tank itself and small pipe network to cause arsenic levels in the small pipe network is very unstable in the process of alternating flow and static state (mainly because of impurities deposited in the pipeline water suddenly brought up and the wall of the scouring action) into consideration, sampling before each cycle begins running as a blank sample.

The test water quality monitoring indicators include: temperature, pH, turbidity, free residual chlorine, alkalinity, hardness, arsenic, total iron, manganese, lead, calcium, magnesium, aluminum, chromium and the particulate matter from July 10, 2017 to October 10 day.

3. Results

As the life of the Eastern Zhou waterworks water supplying is 10 years and the pipes were lay after 2000, which is not such a long time, in its water supply area that the growth ring in metal pipes' wall is not so great. It does not affect the normal pass water capacity of the pipeline. However, the junctions, such as elbows, tees, valves, are with thick growth ring. We can clearly see the rust and there is an apparent precipitation layer after scratch, which is shown in Figure 2 and Figure 3.



Figure 2. Growth ring in pipes' wall



Figure 3. Growth ring cutting

Growth rings can be broadly divided into three layers, corrosion layer, precipitation layer and biofilm layer from the inside to the outside (Md. Rezaul Karim et al. 2010). The outermost growth rings mucosa is biofilm. The growth form of bacteria in the water supply network is suspended growth in aqueous solution and attached growth in growth rings. Since the drinking water make the condition become the oligotrophic growth environment, bacterial is more vigorous in the adhesion of the wall than suspended growth dominantly (Kyung Hwa Cho et al. 2011; Tain-Junn Cheng et al. 2010). The formation of biofilms is a composite body of the micro-organisms, microbial secretions, microbial debris and the adsorbed organic. The intermediate layer is consist of colloidal particles of calcium carbonate and the pipeline after-precipitation with the characters of brown, porous, uneven layer of sediment and easy to be peeled off. When there is a change of flow rate, pH value or water quality in water distribution system, deposition scaling and other phenomena occur and form a part of the growth rings. the inner layer contacted with the pipes' wall directly is a black, hard, corrosion products that contains iron oxide, ferric hydroxide, ferrous sulfide, etc. In addition, there are a lot of pores within the growth rings. Therefore, it is easy to adsorption of certain ions or micelles. Digesting the interception pipeline growth ring sample by U.S. Perkinelmer 5300DV atomic emission spectrometer, the component results of growth rings are shown in Table 2.

Table 2. The component results of growth rings

| NO. | Diameter | As($\mu\text{g/g}$) | Fe(mg/g) | Mn($\mu\text{g/g}$) | Ca(mg/g) | Zn(mg/g) | Al($\mu\text{g/g}$) | Cu($\mu\text{g/g}$) | Pb($\mu\text{g/g}$) |
|-----|----------|-----------------------|---------------------|-----------------------|---------------------|---------------------|-----------------------|-----------------------|-----------------------|
| 1 | DN50 | 16.38 | 0.88 | 27.10 | 0.33 | 4.96 | 138.75 | 4.25 | 411.85 |
| 2 | DN50 | 13.06 | 3.27 | 64.31 | 0.63 | 2.22 | 60.11 | 3.39 | 181.01 |
| 3 | DN50 | 31.95 | 0.94 | 23.05 | 0.47 | 4.69 | 111.62 | 4.15 | 377.37 |
| 4 | DN100 | 21.74 | 0.89 | 22.55 | 0.43 | 4.73 | 90.90 | 3.45 | 385.45 |
| 5 | DN100 | 47.33 | 1.29 | 19.10 | 1.80 | 4.52 | 537.11 | 38.62 | 305.55 |
| 6 | DN200 | 40.68 | 1.03 | 26.04 | 0.63 | 4.57 | 136.00 | 5.17 | 371.30 |
| 7 | DN300 | 40.24 | 5.09 | 72.64 | 0.76 | 0.92 | 61.41 | 6.08 | 84.46 |
| 8 | DN300 | 34.42 | 2.80 | 58.74 | 0.90 | 2.57 | 115.00 | 9.00 | 220.40 |
| 9 | DN400 | 60.60 | 3.27 | 53.58 | 1.09 | 2.37 | 175.63 | 76.49 | 189.26 |
| 10 | DN400 | 84.24 | 4.16 | 75.02 | 0.80 | 3.42 | 74.04 | 6.87 | 126.49 |
| 11 | DN500 | 115.24 | 4.66 | 83.26 | 0.85 | 3.89 | 67.06 | 7.32 | 85.55 |
| 12 | DN500 | 63.79 | 5.17 | 91.50 | 0.91 | 4.36 | 60.08 | 7.77 | 44.61 |

In addition, the samples of growth rings in different diameters were measured stratification and the average result is shown in Figure 5. Therefore, the main factors impact arsenic enrichment in the pipeline are the stratification of the inner wall of the pipes and the diameter. The arsenic enrichment results are mainly due to the adsorption of Fe, and the growth rings of the metal tube, and the respective metal elements occur within similar complexes binding. Various elements attached to each other so that the of the case of enrichment is significant.

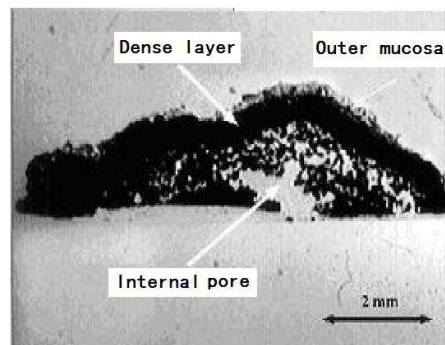


Figure 4. Cross section hierarchical situation of growth ring

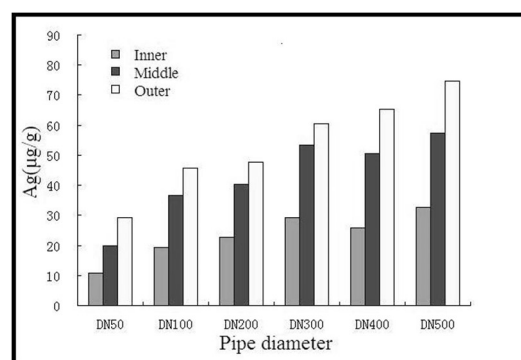


Figure 5. Arsenic enrichment stratified test sample analysis in different pipe diameter

3.1. Velocity of flow

Pipe diameter which is above DN500 is not easy to laboratory tests, so we use DN50 ~ DN400 pipeline for flow test, changing the flow rate is incremented by 0.3m / s to 1.3 m / s, its arsenic release changes are shown in Figure 6.

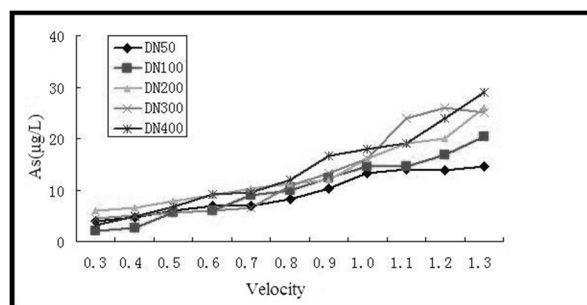


Figure 6. Change law of arsenic release based on the velocity

3.2. pH

The pH test by changing the pH from 7 to 9 and then make hydraulic erosion in DN50 ~ DN400 pipe. When the flow rate increased to an average value of 0.9 ~ 1.1 m/s, or more, the arsenic released is shown in Figure7.

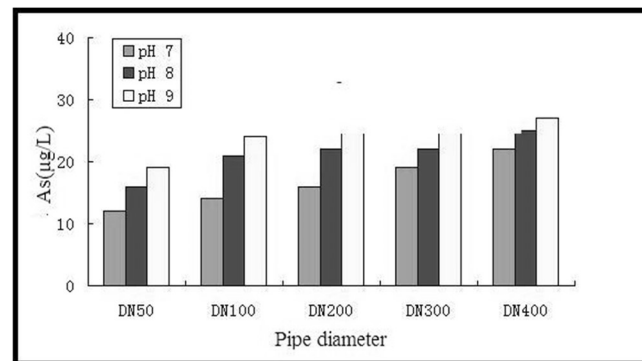


Figure 7. Change law of arsenic release based on the pH

3.3. Particulate matter

Since the test turbidity fluctuation is very small that the particle can't be reflected clearly. The use of the particle counter distribution of particulate matter detection can overcome the above weakness. The average test results are shown in Figure 8 and Figure 9.

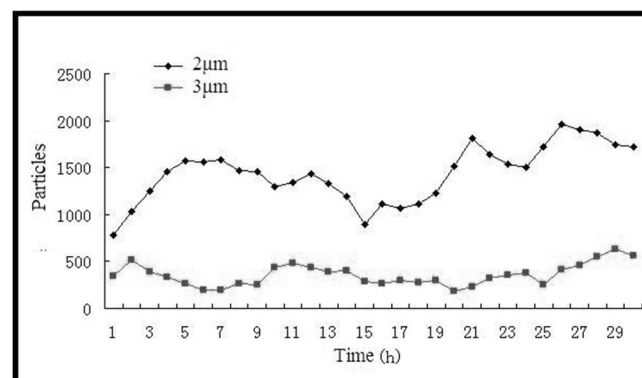


Figure 8. Particles change in washing - arsenic release test (2 µm, 3 µm)

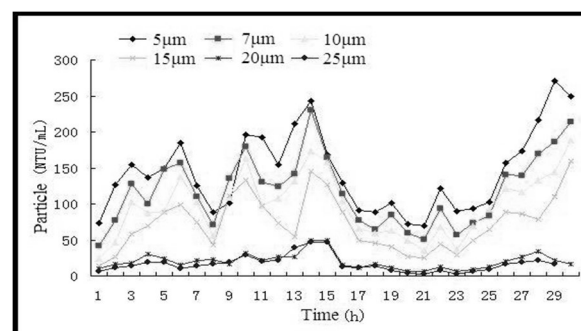


Figure 9. Particles change in washing - arsenic release test (5µm~25µm)

Total particulate matter has got a overall upward trend in the running process, which contrast with turbidity as shown in Figure 10. Water turbidity is fluctuations by not only the volatility of hydraulic but also the sensitivity and stability turbid meter, on the one hand and hydraulic changes. However, both the overall trends are consistent.

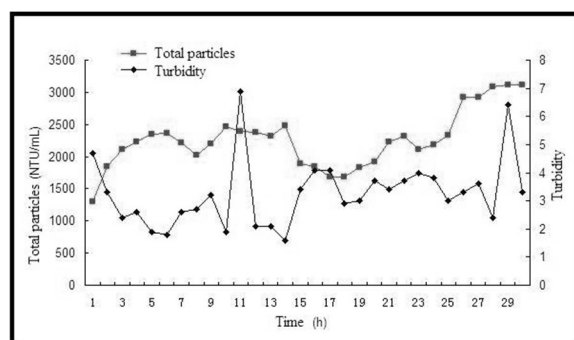


Figure 10. Particles and turbidity

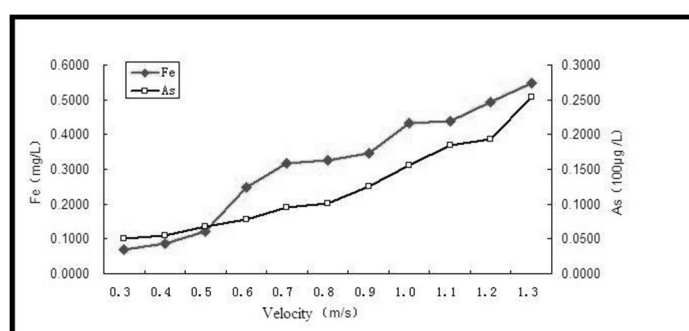


Figure 11. The change rule of arsenic and iron release based on the velocity

Meanwhile, according to the water arsenic concentration study we found a positive correlation between particulate matter and water arsenic concentration.

3.4. Ion

Hydraulic erosion process not only make a release of As but also other metal ions, where significant in Fe and Pb mostly (Suzana C'avar et al. 2005). Take the DN200 metal pipes for example, all metal ions release situation as shown in Table 3 with the flow rate increasing,

Table 3. Release of metal ion

| Flow velocity (m/s) | As (mg/L) | Fe (mg/L) | Mn (mg/L) | Pb (mg/L) | Ca (mg/L) | Mg (mg/L) | Al (mg/L) | Cr (mg/L) | Cd (mg/L) |
|---------------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| Before rinsing | 0.00114 | 0.0895 | 0.02445 | 0.00412 | 18.44 | 2.937 | 0.03234 | 0.00100 | 0.00011 |
| 0.3 | 0.00621 | 0.1580 | 0.02797 | 0.00766 | 19.17 | 3.165 | 0.04503 | 0.00100 | 0.00029 |
| 0.4 | 0.00669 | 0.1767 | 0.03238 | 0.01681 | 19.88 | 3.157 | 0.03424 | 0.00260 | 0.00043 |
| 0.5 | 0.00796 | 0.2113 | 0.03616 | 0.02684 | 18.82 | 2.906 | 0.03077 | 0.00191 | 0.00041 |
| 0.6 | 0.00893 | 0.3387 | 0.03414 | 0.03692 | 19.23 | 3.002 | 0.03090 | 0.00261 | 0.00032 |
| 0.7 | 0.01063 | 0.4076 | 0.03859 | 0.04958 | 19.86 | 3.162 | 0.02718 | 0.00210 | 0.00043 |
| 0.8 | 0.01122 | 0.4151 | 0.04052 | 0.05469 | 20.65 | 3.302 | 0.03342 | 0.00169 | 0.00045 |
| 0.9 | 0.01365 | 0.4354 | 0.04555 | 0.06094 | 20.13 | 3.018 | 0.03249 | 0.00207 | 0.00042 |
| 1.0 | 0.01672 | 0.5232 | 0.04942 | 0.06745 | 20.46 | 3.009 | 0.04227 | 0.00332 | 0.00033 |
| 1.1 | 0.01955 | 0.5275 | 0.05297 | 0.07313 | 20.75 | 3.205 | 0.03403 | 0.00275 | 0.00038 |
| 1.2 | 0.02046 | 0.5829 | 0.05665 | 0.07912 | 21.08 | 3.476 | 0.03998 | 0.00307 | 0.00044 |
| 1.3 | 0.02659 | 0.6380 | 0.05704 | 0.08172 | 21.92 | 4.215 | 0.03792 | 0.00326 | 0.00045 |

The blank sample is water met drinking water standards before rinsing and after a cycle rinse experiments, various types of metal ion release with a generally increasing trend and individuals different. As exceeded standard when in the flow rate increased to 0.7m/s and even 26.59 $\mu\text{g/L}$ in 1.3m/s. Fe exceeded standard when in the flow rate increased to 1.3m/s. Pb exceeded in 0.4m/s and subsequent higher than the standard after continuous washing process; other metal ions without excessive phenomenon.

4. Discussion

Be seen from the table1-3, the arsenic enrichment in the pipe wall range from 10 $\mu\text{g/g}$ (solid sample) to 115 $\mu\text{g/g}$ (solid sample) in a serious condition, which is likely resulting in the release of arsenic contamination in drinking water when flow rate, pH or other the hydraulic water quality conditions change.

At the same time, the content of Zn, Pb and other heavy metal in the inner wall of the pipe is high. Analyze the reasons. For a long-term of immersion scour or inorganic reagents dissolved in water when manufacturing junction the pipeline further adsorption or combined with the formation of precipitation (Tien Vinh Nguyen et al. 2010).

The results showed that, the content of arsenic in the growth rings is decreasing in turn from the outer to the inner. The main reason for the growth ring of the outer layer of mucous membrane adsorption of arsenic is relatively strong, while the middle is a dense layer, less adsorption of arsenic and other ions. Meanwhile, the measured sample results show that the arsenic in pipe segment junction, such as elbows, tees, etc. is higher degree of enrichment significantly.

Most of the pipelines intercepted are laid from 2000 to 2006 except for a small amount of the pipe laid in 1998. The arsenic enrichment significantly increased with the increasing of diameter significantly, while there are few evidence to proofed the relationship between pipe age and arsenic enrichment. In addition, a small amount of households pipes are PVC pipes with a slightly higher growth rings of arsenic enrichment than the metal pipes.

The results show that the arsenic release presents a substantially gentle upward trend with the velocity increment. For pipelines with a diameter of DN100 or smaller, arsenic release phenomenon appears obviously when the flow rate reach 0.8m/s or more, or there is water hammer and eddy. For pipelines with a diameter of DN100 ~ 400, obvious arsenic release phenomenon when the flow rate reach 1.0 ~ 1.3 m/s or water hammer and eddy.

The total concentration of arsenic in water is up to 30 $\mu\text{g/L}$ after scouring test, which make a serious drinking water pollution. Visible, the flow rate of hydraulic scour make great impact on arsenic releasing so that we consider the use of regularly washing for pipes arsenic removal.

The arsenic release aggravated with the increase of pH within the range of 7-9. Total concentration of arsenic can be reached 30 $\mu\text{g/L}$ which shows that arsenic is easier to release in an alkaline environment. And therefore may be considered a method of using the the pH adjusted in pipeline for arsenic removal (Frank Oesterholt, Gerard Martijnse, 2007).

The test results show that the hydraulic erosion and collision become serious with the growth of the flushing time in addition to water eddy current phenomenon, the number of fine particulates (2 μm , 3 μm) is growing. At the same time, the larger particles presents increased at first and then decreased again. Because the scour velocity gradually increases, larger particles crushed by impacting and friction process, then fragmented to a small particle size. (N. Sanjeev Kumar et al. 2010) Thereafter, total number of particles and the density of particles in the water increase. Some small particles are re-gathered and condensed into large particles during the impact, which make a growth on larger particles again.

Previous research had found that the residual As were adsorbed on the inner wall of the pipes' growth rings (J. Theytaz et al. 2009). Fe is the most important component of the growth rings in pipe wall. The positive correlation between both the degree of net release and detailed in Figure 10——logarithmic fit well, $R^2=0.9635$, somewhat less linear fit, R^2 is 0.876. Thus, further shows that the major performance of As residual is related to adsorption of Fe and its compounds in water.

During the test, with the growth of the flushing time, pH and alkalinity have a rising trend. Hardness is 480 ~ 550 (CaCO₃, mg / L), which also shows a slow upward trend, the detection value is slightly exceeded compared to the drinking water health standard; temperature had no significant change.

5. Conclusion

(1) As enrichment in water distribution is mainly due to the adsorption of growth rings to As and binding reaction of other metal to As in pipe wall. And its release mainly comes from the desorption of the solid surface of the Fe salt. pH and other water quality parameters have a certain influence on As release, too.

(2) The As concentration is 3 µg / L or less that no significant As release phenomenon in general survey of peripheral water quality

(3) For the impact of the pipeline properties on the water quality of the distribution, the steel pipe (corrosion) has less than ductile iron pipe. The short pipeline laid age does not affect the water quality of the pipe network in this research.

(4) Hydraulic status has great impact on water quality.

(5) Arsenic enrichment increases from the outer to the inner in a descending order in pipeline growth rings and relationship between arsenic enrichment and pipe laying age is not obvious.

(6) Arsenic release is aggravated with the increase of pH within the range of 7~9.

(7) Particulate matter and water turbidity is basically the same with a positive correlation with water arsenic concentration both.

(8) Pipe erosion-arsenic release experiments, in addition to As, Fe and Pb also showed significant release phenomenon. As and Fe have a positive correlation between the degree of net release.

(9) Based on the debris of accumulation, select the unidirectional flushing method, high-pressure water jet, scraping tube gas pulse cleaning method for pipe wall cleaning, so as to achieve the purpose of the removal of the pipe network residual arsenic.

Acknowledgements

This research is supported by 2018 Guangxi Vocational Education Reform Research Project: Research and Practice of Innovation and Entrepreneurship Education Model in Higher Vocational Colleges Based on "Five Industry Integration". (ID: GXGZJG2018B043).

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