

# Would Use of Contaminated Water for Irrigation Lead to More Accumulation of Nitrate in Crops?

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## Abstract

Nitrate content in agricultural crops is of interest to governments and the general public owing to the possible implications for health. There are two pathways for nitrate to enter into human body: food and drinking water. The dietary intake of nitrate is usually much larger than that from drinking water. This work investigated nitrate content in various crops and its relevance to the nitrate concentration of irrigation water in an arid region, Northwest China. It revealed that irrigation water with high concentration of nitrate tends to produce crops with high build-up of nitrate. Nevertheless, all sampled crops adjust their nitrate distributions in a way that the edible parts contain much less nitrate except lettuce. The nitrate content in the edible root of lettuce reached up to 5900 mg/kg exceeding the limit set by the European Commission. The findings, although preliminary, supplemented the existing literature and raised questions for further study.

**Keywords:** nitrate, distribution in crops, wastewater, arid region

## 1. Introduction

Water use has been growing globally putting unprecedented pressure on renewable, but finite water resources, especially in water-stressed regions. An increasing number of rivers now run dry before reaching the sea for substantial periods of the year. In many areas, groundwater is being pumped at rates that exceed replenishment, depleting aquifers and the base flows of rivers (Postel, 2000). In the future, climate change and bio-energy demands are expected to amplify the already complex relationship between world development and water demand. According to the WHO reports that within the next 50 years, more than 40% of the world's population will live in countries facing water stress or water scarcity (WHO, 2006). At the same time, the production and discharge of domestic wastewater is rapidly increasing in developing countries due to population growth, urbanization, and economic development. The produced municipal wastewater in China increased from 23.03 ( $10^9$  m<sup>3</sup>/yr) in 2002 to 35.48 ( $10^9$  m<sup>3</sup>/yr) in 2009. Despite billions invested in improving wastewater management worldwide, Ujang and Henze (2006) pointed out that 95 per cent of wastewater generated enters the environment without proper treatment. In developing countries, wastewater treatment plants represent one of the major investments. However, restricted local budgets, lack of local expertise, and lack of funding resulted in inadequate operation of wastewater treatment plants in developing countries (Paraskevas et al., 2002).

Due to the increasing trend in both water demand and wastewater discharge and the high cost of treatment facilities, there is an increasing use of partially treated and untreated wastewater in irrigated agriculture in developing countries. Indeed, the use of domestic wastewater for crop production has been practiced for several centuries in one form or another in countries such as China, India, Australia, Germany and UK. However, the importance of wastewater reuse in relation to sustainable development was recognized in recent decades. The question about from where the extra water for further development is to come, has led to a scrutiny of present water use strategies. It appears that reuse of wastewater is a key toward rational and efficient use of available water.

Benefits of wastewater use in agriculture may include: reliable and low cost source of water that is available to farmers whenever they need it (unlike canal irrigation) even in the dry season; nutrients present in wastewater may replace chemical fertilizers saving a lot of money to farmers and increasing crop yields as well. Pescod (1992) estimated that typical wastewater effluent from domestic sources could supply all of the nitrogen and much of the phosphorus and potassium that are normally required for agricultural crop production. Therefore, it

can be considered as a means of reducing poverty for a large portion of people in developing countries. Applying wastewater to land also limits the pollution of rivers, canals and other surface-water resources, which would otherwise be used as disposal outlets. From the environmental engineering point of view it is of interest that agricultural fields can function as a tertiary treatment step through using effluents in irrigated agriculture. Besides, using wastewater means that there is less demand for fresh water for irrigation.

On the other hand, use of wastewater irrigation may result in serious environmental problems, contaminating groundwater with nitrates and heavy metals (especially if the wastewater contains industrial wastes). Such pollutants may build up over time in the soil and be taken up by food crops. Therefore, policymakers need to develop better understanding on the consequence of using wastewater and comprehensive strategies for managing wastewater tailored to local socioeconomic and environmental conditions. It must be assured that short-term benefits of wastewater irrigation are not offset by health and environmental costs. In this work, the food safety issue in relation to the use of wastewater was examined from the angle of nitrate accumulation in crops.

Nitrate ( $\text{NO}_3^-$ ) is an integral part of the nitrogen cycle in the environment. It is oxidized form of nitrogen and highly soluble in water. It is invisible, odorless and tasteless. Nitrate is commonly found in soil and water and is the most effective form by which plants obtain their nitrogen. However, it is also most readily lost from soil by leaching in drainage water, which may be considered as one of the reasons for over-application of nitrogen fertilizers (Addiscott, 2005; Razowska-Jaworek & Sadurski, 2004). A particular concern of nitrogen use in agricultural practices is the nitrate contamination in waters and accumulation in agricultural products. The primary reason for the concern is the threat it poses to human health. The toxicity of nitrate to humans is mainly attributable to its reduction to nitrite. The major biological effect of nitrite in humans is its involvement in the oxidation of normal Hemoglobin (Hb) to met-Hemoglobin (met-Hb), which is unable to transport oxygen to the tissues. The Hb of young infants is more susceptible to met-Hb formation than that of older children and adults. Drinking water with elevated nitrate levels has been highlighted as the cause of the so-called "blue baby syndrome" (Majumdar, 2003). It is also associated with respiratory and reproductive system illness, some cancers, and thyroid problems (Dutt & Lim, 1987; Moore, 2003). Study by Parslowa et al. (1997) indicated a positive relationship between the incidence of childhood-onset insulin-dependent diabetes mellitus and levels of nitrate in drinking water. On the other hand, studies have shown that vegetables eaten by people contribute about 72-94% of the total daily intake of nitrate (Dich et al., 1996; Eichholzer & Gutzwiller, 1998). De Martin and Restani (2003) showed that leafy green vegetables accumulate high amounts of nitrates, concentrations reaching up to 6000 mg/kg. Petersen and Stoltze (1999) surveyed the contents of nitrate and nitrite in lettuce, leek, potato, beetroot, Chinese cabbage and white cabbage on the Danish market. The highest content of nitrate was found in lettuce followed by beetroot and Chinese cabbage. Although previous studies have greatly improved the understanding on the possible range of nitrate contents in various agricultural products, the knowledge of nitrate distribution within crops is still limited. Santamaria et al. (2001) showed that the nitrate content in vegetable organs can be listed in the decreasing order from petiole, leaf, stem, root to seed. Nevertheless, it is not clear if the decreasing order from stem, root to seed also holds for tall crop plant such as corn. Factors such as plant species, soil, nitrogen fertilization, light, climatic conditions and water availability affect nitrate accumulation.

The present study examined the nitrate distribution in various crops. The objective is to further improve the understanding on the health risk of nitrate contamination of crops by differentiating its concentration in edible parts from non-edible parts and highlighting its relevance to irrigation water. It is hypothesized that irrigation water quality is another fact influencing crop nitrate accumulation. The rationale is that different irrigation water resources may contain different levels of nutrients and have different ranges of water temperature which might affect the nitrate intake of crops and nitrate translocation inside crops as well. By choosing survey sites within the same climate zone having similar soil condition, and farming methods, the difference in nitrate contents of crops among survey sites, if detected, may be attributed to the difference in irrigation water quality.

## 2. Method

The middle reaches of the Heihe River was chosen as the study area. Heihe River is the second largest inland rivers in China. Its main stream, with a length of 821 km, originates from the Qilian Mountains of Qinghai Province, flows through the Zhangye Basin, which is part of the ancient Silk Road, and ends up in the Inner Mongolia Autonomous Region. The catchment of the middle reaches, or the Zhangye Basin, covers an area of  $1.08 \times 10^4 \text{ km}^2$  extending from  $38^\circ 30'$  to  $39^\circ 50'$  N and  $99^\circ 10'$  to  $100^\circ 52'$  E. Along the main stream, the middle reaches starts from the Yingluo Gorge and ends at the Zhengyi Gorge (Figure 1).

Zhangye Basin is characterized by a dry continental climate, with mean annual precipitation of about 200 mm,



measured. For vegetables, leaf and root were measured separately. Besides, the nitrate and ammonium ( $\text{NH}_4^+$ ) concentrations of irrigation water entering into the farmland were measured with the Horiba compact nitrate ion meter (LAQUAtwin-B743) for water and HACH Portable Meter (DR/890), respectively. Furthermore nitrate concentrations in drinking well water were also tested where the nitrate concentrations in crops were measured. The field investigation was conducted from August 8 to August 18, 2012 and all measurements were done under fine weather during daytime. A conventional approach to study nitrate accumulation in crops is to conduct split plot experiments under well-controlled conditions. Such kind of study is useful for better understanding the fundamental mechanisms related to nitrate accumulation in crops. However, this approach does not fit the present study because the objective of this work was to grasp the actual situation about nitrate contamination of market ready crops in the target area. Besides, agricultural products sold in markets were not used in this study because the storage process may alter the nitrate contents and its relevance to irrigation water could not be discussed.

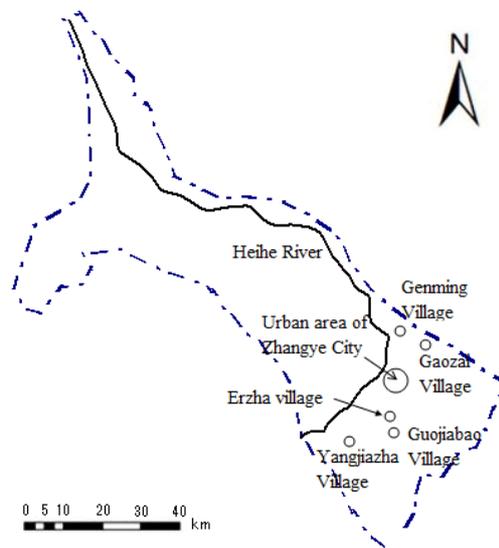


Figure 2. Zhangye Basin and sampling sites

### 3. Results and Discussions

#### 3.1 Wastewater-Fed Field

At the two sites where sewage-dominated water was used for irrigation, the concentrations of nitrate in water entering into corn fields were found to be 20 mg/L in the Gaozai village and 110 mg/L in the Genming village, respectively at the time of sampling. And the concentrations of ammonium ( $\text{NH}_4^+$ ) in the water were also tested at the same time and they were 6 mg/L and 0.2 mg/L, respectively. In the Gaozai village, we randomly selected two corn plants; one mature and another immature. The maturity is judged by the size and color of corn ears. The mature corn ear was 20 cm long and golden color while the immature was 10 cm long and pale green color. However, in the Genming village, immature corn was not spotted at the time of survey.

Figure 3 shows the nitrate concentration distributions along the corn stalks. The distributions along the mature corns from both sites showed very similar pattern. The nitrate concentration was as high as 3700 mg/kg at the brace roots but decayed quickly along the stalk. In the immature plant, the nitrate concentration at the bottom was not significantly higher than the top. The maximum concentration occurred somehow in the middle of the plant where a single ear grew. The nitrate concentrations in niblets of both the mature and immature corns were 110 mg/kg and 210 mg/kg, respectively. This comparison suggests that the corn plant adjust its nitrate build-up through its growing period in a way that the edible part contains the least nitrate at the end. In other words, the final distribution of nitrate in corn plant is in favor of human beings. Besides, the relatively low concentration of nitrate at the brace roots of immature corn plant implies that the plant accumulated more nitrate or assimilated less nitrate at its late growing stage. This provides a hint for better nitrogen fertilizer application. The nitrate concentrations in the drinking well in Gaozai village was also tested and found to be 68 mg/kg, exceeding the WHO standard but not Chinese standard. Although evidence is not available at this stage of investigation to

assert that the drinking water (groundwater) contamination of nitrate is related to the use of wastewater in irrigation, it is a concern that should be addressed and clarified.

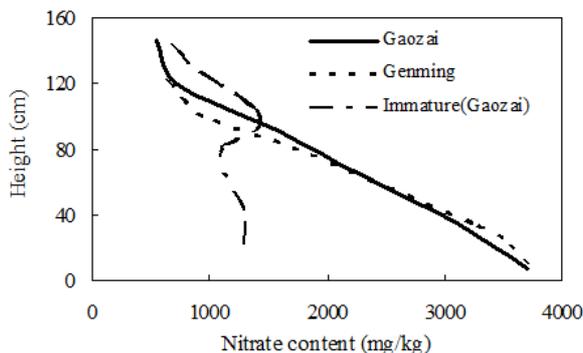


Figure 3. Nitrate distributions in wastewater-irrigated corn stalks

### 3.2 Groundwater-Fed Field

In the Guojiabo and Erzha villages, the groundwater is used to irrigate vegetable farmlands. The two villages are about 1.5 km apart and their irrigation water was taken from wells about 30 m deep. Cauliflower, broccoli, lettuce, beet, tomato, Chinese cabbage, green pepper, red pepper, lantern chilli, cucumber were sampled for nitrate concentration. The measurement results are compiled in Figure 4.

The nitrate concentration in broccoli leaf was as high as 5300 mg/kg. However, the nitrate concentrations in broccoli thick stalk and flower head were 2600 mg/kg and 860 mg/kg, respectively. Cauliflower and Chinese cabbage contained high amount of nitrate in leaf but much less in edible part. Among all measured, lettuce was the only one having concentrations higher than 3000 mg/kg in both leaf and root. Beet leaf and root had quite the same but moderate amount of nitrate. It should be noted that both the leaf and root of lettuce are consumed as food by humans. For broccoli, fleshy flower head and the top part of thick stalk are edible. Beet is now usually used for making sugar. The distinctive patterns of nitrate distribution within Lettuce and Beet revealed in this study may serve as a call for further investigation on nitrate accumulation mechanisms in crops. Besides, water quality test showed that the nitrate concentrations in the drinking well water was 78 mg/kg in the Guojiabo village and as high as 160 mg/kg in the Erzha village. This suggested that nitrate contamination of groundwater has progressed in the vegetable-growing villages.

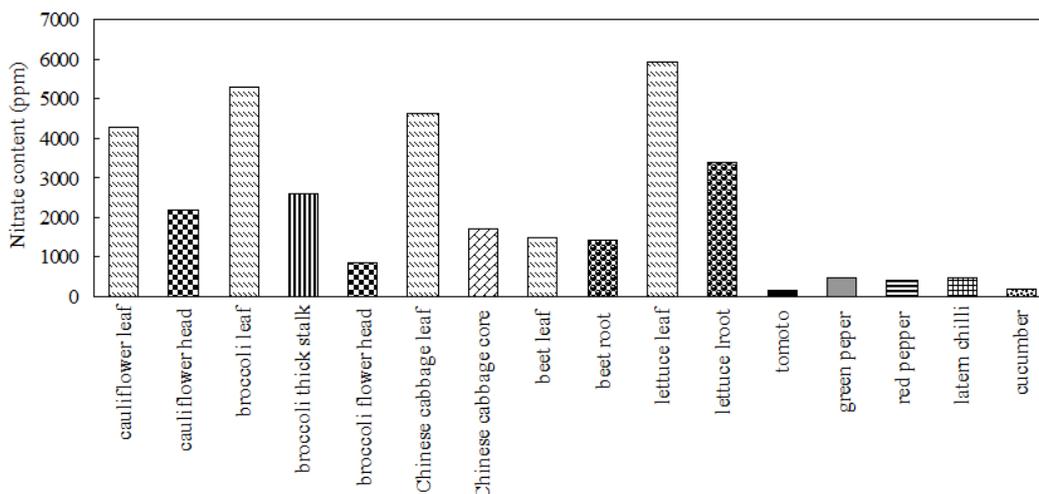


Figure 4. Nitrate contents in various agricultural products

### 3.3 River Water

In the Yangjiazha village where the water from the Heihe River is used for irrigation, the nitrate concentration in the irrigation water was tested to be 15 mg/l. Figure 5 shows the nitrate distribution along a corn plant sampled from this village. The corn plant in this river-water-irrigated area shared similar characteristics with that in the wastewater-irrigated area in terms of the vertical decreasing pattern. However, the nitrate concentration in the river-water-fed plant decreased vertically much faster than the wastewater-fed plant. Therefore, the total build-up of nitrate is much less when river water is used for irrigation. In the Yangjiazha village, the nitrate concentration of the drinking well water was 34 mg/kg, below the WHO standard.

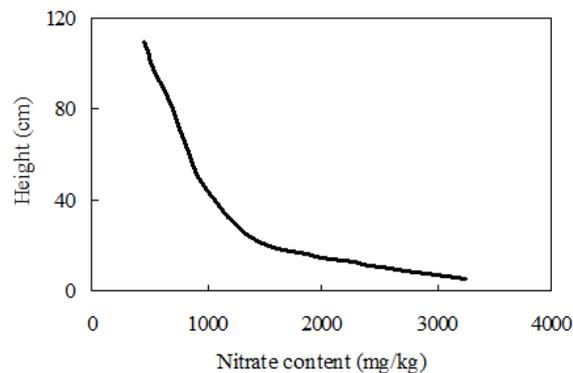


Figure 5. Nitrate distributions in river-water- irrigated corn stalk

### 3.4 Discussion

Prior to the work of Santamaria et al. (2001), other studies (Maynard et al., 1976; Ostrem & Collins, 1983) also reported that leaf blades have a lower nitrate content than stems and petioles, and young leaves show a lower nitrate concentration than older leaves. The present study improved the understanding on nitrate distribution in vegetables by showing that there could be a difference between edible and non-edible parts. Examining the nitrate distribution in such a manner is important from the food safety perspective. Such a classification and corresponding knowledge may help develop guideline for food processing. Besides, the vertical profiles along corn stalks obtained from this study supplemented the literature with regard to nitrate distribution in high plants.

The reason for more accumulation of nitrate in corn stalks irrigated by contaminated water could not be explained by the available data from the current study. However, a hypothesis is that it might be influenced by water temperature because rates of nutrient uptake and internal transport mechanisms could be considered as temperature-dependent. Since the river water originated from snow-melting, the preliminary field data found that the river water temperature was 2~3 °C lower than the sewage-dominated irrigation water.

Plants have the ability to maintain an internal environment with a composition different from that of their surroundings. The internal environment (chemical contents) of the plant body remains more or less constant whereas the outside environment is highly variable. Whenever an ion moves into or out of a cell unbalanced by a counter ion of opposite charge, it creates a voltage difference across the membrane called electrogenic pump. A fundamental expression for characterizing such a system is the Nernst potential  $E$  as below:

$$E = (RT/zF) \ln(C_o/C_i) \quad (1)$$

Where  $R$  is the gas constant,  $T$  is the absolute temperature,  $z$  is the charge of ion, and  $F$  is the Faraday constant.  $C_o$  and  $C_i$  are the ion concentrations outside and inside the cell, respectively.

Therefore, nutrient uptake by plant is temperature-affected in general although the sensitivity might be species-dependent.

In river water-irrigated fields, fertilizer is applied into soil so that nutrients are uptaken by plant roots. In wastewater-fed fields, flood irrigation is the normal practice so that nutrients may also be absorbed via stalk due to the direct contact with nutrients-rich wastewater.

Because water temperature measurements at different locations were not done continuously over a sufficiently long period of time, and plant growth was not simultaneously monitored with water temperature, quantitative

evaluation of water temperature effect on nutrient uptake was not possible at this stage. Nevertheless, the preliminary findings highlighted the need for further study to quantify the true difference in water temperature regime among different irrigation waters and analyze its effects on nutrient uptake and vertical transport.

### 3.5 Statistical Analysis

In total, 10 mature corn plants were sampled in the Zhangye Basin in August 2012. The concentrations of root part varied from 3300 mg/kg to 3700 mg/kg with the mean and standard deviation being 3588 mg/kg and 118 mg/kg, respectively. Meanwhile, the concentrations of top part varied from 450 mg/kg to 770 mg/kg with the mean and standard deviation being 624 mg/kg and 132 mg/kg, respectively. Moreover, the concentrations of corn nib lets varied from 96 mg/kg to 210 mg/kg with the mean and standard deviation being 146 mg/kg and 41 mg/kg, respectively. To confirm that the observed difference in nitrate content connotes any real difference between wastewater-fed and river-water-fed groups, the test of significance should be conducted. For large samples, it can be done by a Z-test. Since the sample size is limited in the present study, the significance test was conducted by the so-called change-point analysis, which is a non-parametric method. It is an effective and powerful statistical tool for detecting mean shifts in a time series. The previous applications of change point detection methods include bio-informatics applications (Erdman & Emerson, 2008), network traffic analysis (Kwon et al. 2006), climatology (Reeves et al., 2007) and oceanography (Killick et al., 2010). The procedure constructed by Taylor (2000) for performing a change-point analysis iteratively uses a combination of cumulative sum charts (CUSUM) and bootstrapping without replacement. To use this technique under the context of this study, nitrate concentrations of corns measured in sewage-irrigated fields were appended by concentration data taken from river-water-irrigated fields to form sequential datasets for different parts of corn. Then, the change-point analysis was applied to datasets to detect any significant change in data. If a significant shift in mean appeared between sewage-related and river-related data, the null hypothesis of no difference in nitrate content between wastewater-fed and river-water-fed corns could be rejected. As shown in Figure 6, for the middle part of corn, there was a significant shift in mean concentration of nitrate from wastewater-fed to river-water-fed groups. However, for the root and top parts and niblets of corn, change-point analysis revealed that the null hypothesis of no significant difference in nitrate content between wastewater-fed and river-water-fed groups was retained.

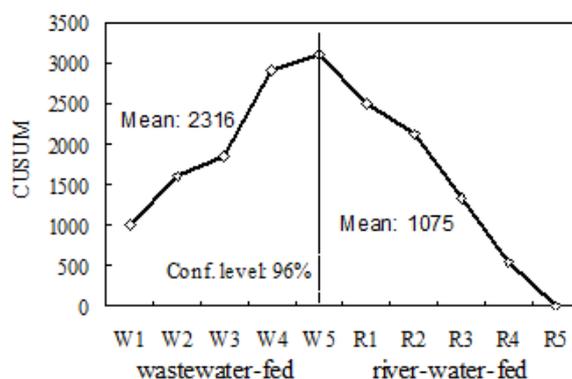


Figure 6. Test of significance by change-point analysis for the nitrate content in the middle part of corn plant

## 4. Conclusions

By looking into the difference in nitrate content among agricultural crops grown with different types of irrigation water, it revealed that the accumulation of nitrate in crops is affected by the type of irrigation water. The use of sewage water for irrigation due to water scarcity may cause more nitrate build-up in crops. In particular, high concentration of nitrate was found in the middle portion of corn stalk when irrigated with wastewater. Such a finding has never been reported before and it was hypothesized to be attributable to water temperature. Nevertheless, by measuring nitrate contents in different parts of various agricultural crops, the present work further revealed that even irrigated with contaminated water, the edible parts of various crops such as corn niblets, broccoli flower head are much less contaminated compared with non-edible parts such as corn stalk and Chinese cabbage leaf. The nitrate in lettuce and beet was found to have quite uniform distribution, but nitrate

content in seed-corn plant can be listed in the decreasing order from brace roots, stalk to niblets. Among all sampled, lettuce is the only one having very high concentration in edible parts. Therefore, it can be stated that many crops adjust their nitrate content distributions in favor of humans.

Although, the use of low quality water for irrigation may not cause significant contamination of edible parts of crops, it can pollute groundwater to the extent that it is no longer suitable for drinking purpose. Therefore, integrated management is required to minimize potential negative effects of wastewater reuse.

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