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# Occurrence and exposure evaluation of perchlorate in indoor dust and diverse food from Chengdu, China

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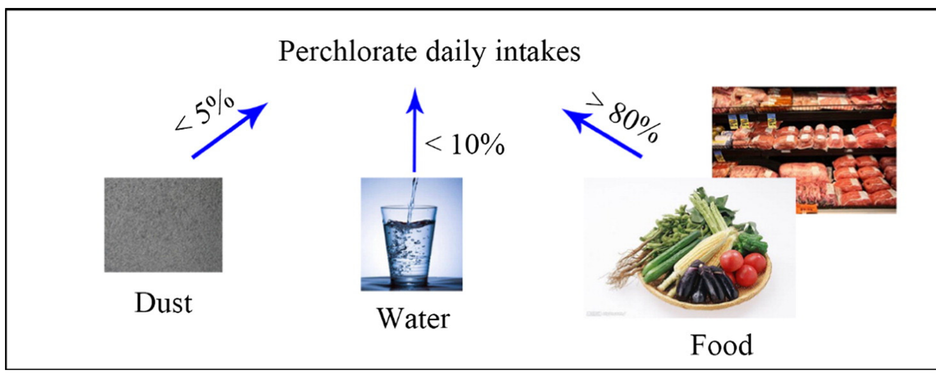
## HIGHLIGHTS

- High perchlorate levels were found in the indoor dust and food samples.
- Indoor dust contributed little to the daily perchlorate intake.
- Food consumption was the primary perchlorate exposure route to Chengdu people.
- The daily mean perchlorate intake was below the US EPA reference.

## GRAPHICAL ABSTRACT

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## ABSTRACT

A total of 688 samples, including 520 food samples belonging to 29 food types in 10 groups, 63 drinking water, and 105 indoor dust samples were collected during May to November in 2014 in Chengdu and Tianjin (only dust samples in Tianjin), China to investigate the perchlorate levels in these samples and to estimate the related exposure to Chinese people. Significant difference in indoor dust perchlorate levels was found between Chengdu and Tianjin, with the concentrations ranging from 0.11 to 38.8 mg/kg in Chengdu, and from 0.72 to 119 mg/kg in Tianjin. The mean perchlorate levels in 10 groups of food samples were in the order of vegetables > wheaten flour ≈ egg, and wheaten flour > milk > sea food > livestock meats ≈ poultry ≈ freshwater fish > fruits > steamed rice, while no statistical difference in perchlorate levels was found between egg and milk. Exposure evaluation indicated that indoor dust contributed little (less than 5%) to the total daily perchlorate intake, and food consumption was the primary perchlorate exposure route for Chengdu people, followed by drinking water. Based on the median perchlorate levels obtained in this study, the daily perchlorate intake was below the reference does suggested by the US EPA.



## 1. Introduction

Despite that perchlorate can originate naturally (Urbansky et al., 2001; Dasgupta et al., 2005), anthropogenic sources are still recognized as the primary source for perchlorate in the environment, because perchlorate is widely used in many industrial and personal products,

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e.g., solid rocket propellants, munitions, fireworks, roadside flares, air bag inflation systems, and pharmaceutics (Urbansky, 1998; Zewdie et al., 2010). Recently, perchlorate has raised more concerns due to its persistence and ubiquitous occurrence in aquatic environment and drinking water, with concentrations ranging from below 0.1 to 2300 ng/mL (Kosaka et al., 2007; Kannan et al., 2009; Wu et al., 2010; Qin et al., 2014). In addition, perchlorate was found in human blood, saliva, breast milk, and urine, with concentrations ranging from below 0.1 to 160 ng/mL (Kirk et al., 2005; Blount et al., 2006; Kannan et al., 2009; Zhang et al., 2010b). Previous study has documented that perchlorate can inhibit iodide uptake by the sodium iodide symporter (NIS), which could result in thyroid dysfunction (Wolff, 1998). Based on the study by the U.S. National Academy of Science, a reference dose (RfD) of 700 ng/kg/day has been established by the U.S. Environmental Protection Agency (US EPA, 2005; Huber et al., 2010).

In addition to the aquatic environment, perchlorate was found in soil samples with concentrations ranging from 1 μg/kg to 216 mg/kg (Rao et al., 2007; Jackson et al., 2010; Ye et al., 2013; Gan et al., 2014). Previous studies indicated that leafy plants could accumulate perchlorate from contaminated soil or irrigation water (Jackson et al., 2005; Sanchez et al., 2005; Voogt and Jackson, 2010). A number of investigations suggested that perchlorate exists in diverse food from the U.S. (Murray et al., 2008; Huber et al., 2010), Canada (Wang et al., 2009), and Korea (Lee et al., 2012), including vegetables, fruits, dairy products, meats, fishes, and eggs, at concentrations up to 536 ng/g ww (wet weight) with the detection rate around 60%. Based on the total diet study and biomonitoring data, Huber et al. (2010) suggested that food is the predominant exposure way to the U.S. people. However, to date, little is known regarding the perchlorate levels in vegetables and other agricultural products in China. Only two studies documented that perchlorate existed in milk and crops, at concentrations up to 9.1 ng/mL and 4.88 ng/g dw (dry weight), respectively (Shi et al., 2007; Ye et al., 2013). Our previous study indicated that perchlorate was widely detected in soil samples from China, at concentrations up to 216 mg/kg (Gan et al., 2014). The high perchlorate levels in soil may cause potential risk through trophic transfer of perchlorate from soil to higher organisms via plants. Therefore, the investigation of perchlorate levels in the agricultural products and its contribution to daily perchlorate intake to Chinese people is necessary.

Quite recently, Wan et al. (2015) and Gan et al. (2014) found high levels of perchlorate in indoor and outdoor dust samples from China, at concentrations up to 104 and 5300 mg/kg, respectively. The perchlorate levels in the indoor dust from China were 4 to 60 times higher than those in other countries (Wan et al., 2015). This original study revealed a new perchlorate exposure way to human, although the contribution of the dust to the total perchlorate exposure was less than 5%. Hence, potential risk of high levels of perchlorate in the indoor dust still merits further investigation.

Therefore, the primary objectives of this study were to investigate the perchlorate concentrations in diverse food from Chengdu, China, including vegetables, fruits, milk, eggs, meats, fishes, sea food, and crops, and to estimate the related exposure to Chinese people. The perchlorate levels in the indoor dust and drinking water (both tap water and bottled water) were studied in parallel to reveal the contributions of different exposure pathways to Chinese people. To our knowledge, this is the first report regarding the levels of perchlorate in various agricultural products in China.

## 2. Materials and methods

### 2.1. Materials

Perchlorate was purchased from Sigma-Aldrich (St. Louis, MO, USA). The $^{18}$O-labeled perchlorate used as internal standard (IS) was obtained from Cambridge Isotope Laboratories (Andover, MA, USA). HPLC-grade methanol and acetonitrile were obtained from J.T. Baker (Phillipsburg, NJ, USA) and CNW Technologies GmbH (Germany), respectively. CNW BOND carbon-GCB (50 mg, 1 mL) cartridges were used for sample clean-up. Milli-Q water was used throughout the study. All other reagents used in this study were of HPLC or analytical grade.

### 2.2. Sample collection

A total of 520 food samples belonging to 29 food types, 12 bottled water samples, and 51 tap water samples were collected during September to November in 2014 (Table 1). All of the samples except sea food were randomly purchased from local markets that are located in western, eastern, northern, and southern regions of Chengdu. Chengdu is the provincial capital of Sichuan province, China, and is the largest city in the southwestern China. Chengdu is the home of panda, it currently has a population of 14.2 million, and has a large volume of trade on agricultural products with many neighboring provinces. Sea food samples were obtained from the largest sea food market of Chengdu, which provides approximately 80% sea food to the local residents.

Indoor dust samples (n = 51, D1–D51, Table S1) were collected in Chengdu during July to August in 2014. Settled dust in bedrooms and living rooms of homes was sampled using a hand-held brush and a paper, and sealed in a glass bottle which was precleaned with a solution of ethanol containing 5% (w/w) NaOH and Milli-Q water, respectively. While tap water samples were collected from the same home in Chengdu. Chengdu is a typical inland city in southwestern China, the annual rainfall is around 1000 mm. To compare the difference of perchlorate levels

**Table 1**
Perchlorate levels in the drinking water and food samples in Chengdu city, China (ng/g for food samples and ng/mL for milk and drinking water).

| Food group | | n | Mean | Min | Max | Median | Food group | | n | Mean | Min | Max | Median |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Fruits | Apple | 20 | 5.46 | n.d. | 28.9 | 1.92 | poultry | Chicken | 20 | 6.24 | n.d. | 15.5 | 5.79 |
| | Orange | 20 | 3.64 | 0.58 | 14.7 | 2.57 | | Duck | 20 | 3.83 | n.d. | 12.3 | 3.73 |
| | Pear | 20 | 2.90 | n.d. | 17.0 | 2.03 | freshwater fish | Crucian carp | 20 | 4.45 | n.d. | 15.7 | 4.04 |
| | Banana | 20 | 4.82 | LOQ | 22.2 | 1.92 | | Common carp | 20 | 4.48 | n.d. | 16.0 | 3.66 |
| Light vegetables | Cabbage | 20 | 33.2 | 2.94 | 216 | 21.7 | | Grass carp | 20 | 4.43 | 1.40 | 11.7 | 3.40 |
| | Potato | 20 | 3.50 | LOQ | 12.0 | 2.13 | | Spotted silver carp | 20 | 4.13 | n.d. | 12.4 | 4.20 |
| | Radish | 20 | 10.5 | 0.74 | 47.2 | 4.86 | sea food | Hairtail | 5 | 5.94 | 3.17 | 8.80 | 5.52 |
| | Cucumber | 20 | 12.0 | n.d. | 114 | 5.47 | | Mantis shrimp | 5 | 12.1 | 6.89 | 15.8 | 13.4 |
| Brunet vegetables | Lettuce | 20 | 25.1 | 9.31 | 73.2 | 20.2 | | Greasy back shrimp | 5 | 4.98 | 3.64 | 7.02 | 4.72 |
| | Spinach | 20 | 156 | 53.9 | 417 | 141 | | Scallop | 5 | 6.34 | 4.40 | 8.25 | 5.63 |
| | Tomato | 20 | 2.92 | 0.53 | 8.29 | 2.19 | | egg | 20 | 15.3 | LOQ | 22.5 | 17.7 |
| | Eggplant | 20 | 5.00 | 0.53 | 21.5 | 2.71 | | milk | 20 | 14.4 | 9.38 | 25.2 | 12.8 |
| | Carrot | 20 | 26.2 | n.d. | 97.9 | 19.3 | | Steamed rice | 20 | 3.25 | n.d. | 9.05 | 3.39 |
| Livestock meats | Pork | 20 | 4.60 | n.d. | 13.8 | 4.28 | | Wheaten flour | 20 | 17.6 | 7.04 | 29.9 | 17.5 |
| | Beef | 20 | 5.61 | n.d. | 28.8 | 4.22 | drinking water | Tap water | 51 | 0.86 | 0.57 | 1.61 | 0.83 |
| | | | | | | | | Mineral water | 12 | 1.03 | LOQ | 2.84 | 0.75 |

n.d.: not detected; LOQ: below the limit of quantification.