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OPEN Iron-fortified water: a new approach for reducing iron deficiency anemia in resource-constrained settings

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A new approach for fortification of drinking water is presented for combating iron deficiency anemia (IDA) worldwide. The idea is to leach Fe from a bed containing granular metallic iron (Fe⁰), primarily using ascorbic acid (AA). AA forms very stable and bioavailable complexes with ferrous iron (Fe^{II}). Calculated amounts of the Fe^{II}-AA solution can be added daily to the drinking water of households or day-care centers for children and adults (e.g. hospitals, kindergartens/schools, refugee camps) to cover the Fe needs of the populations. Granular Fe⁰ (e.g., sponge iron) in filters is regarded as a locally available Fe carrier in low-income settings, and, AA is also considered to be affordable in low-income countries. The primary idea of this concept is to stabilize Fe^{II} from the Fe⁰ filter by using an appropriate AA solution. An experiment showed that up to 12 mg Fe can be daily leached from 1.0 g of a commercial sponge iron using a 2 mM AA solution. Fe fortification of safe drinking water is a practicable, affordable and efficient method for reducing IDA in low-income communities.

Iron deficiency is reported to be the most prevalent nutritional deficiency worldwide, affecting about 5 billion people¹⁻⁴. Iron deficiency is the main cause of anemia (iron deficiency anemia—IDA) in children and women^{4,5}. The prevalence of anemia is five times higher in low-income countries than in high-income countries^{6,7}. Another important reason for this high prevalence is that infectious diseases such as bacterial infections, hookworms, human immunodeficiency virus or acquired immunodeficiency syndrome (HIV/AIDS), intestinal parasitic infections, malaria, and tuberculosis are common conditions associated with a reduction of blood constituents, including iron^{1,2,4}. Iron deficiency impairs cognitive development in children, reduces work capacity in adults, increases the risk of morbidity, induces poor pregnancy outcomes, and impairs immunity^{1,3,4,8}. Iron deficiency is considered to be one of the most expensive diseases in the world, according to the World Health Organization (WHO)^{3,4,8-11}. This is because IDA induces economic losses in the form of (1) cognitive losses among children, (2) lower productivity among adults, and (3) associated increased medical expenses for all groups^{9,11}. Fortunately, IDA is preventable and curable, and doing so is a priority for the WHO^{10,11}. However, despite concerted efforts for iron supplementation (e.g. beverages, food fortification, injections, syrups, drugs, pills, salt fortification, tablets), not much progress has been realized in the past three decades^{3,4,7,12-16}. IDA maintains its top position as the leading cause of years lived with disability, and still contribute to up to 120,000 maternal deaths per year^{3,11,17}.

Many strategies to prevent and treat IDA have been developed over the years^{2,4,7,12,18-20}. Table 1 gives an overview of the used tools based on the oxidation-state-dependent characteristics of the used iron sources (Fe⁰, Fe^{II}, and Fe^{III}). The four most commonly used strategies are: (1) dietary diversification, (2) food fortification with Fe, (3) multi-micronutrient supplementation, and (4) treatment of parasitic infections contributing to iron deficiency^{2,3,5}. A fifth strategy that has been reported only in Brazil, is drinking water fortification with soluble ferrous iron (Fe^{II})^{7,12,21-24}. In this effort, a favored short-term and cost-effective strategy that is relatively easy to implement at large scale, is the use of fortification programs at kindergartens and schools⁵. For the prevention of IDA, the WHO recommends a daily iron supplementation for three consecutive months in a year to all premenopausal women, adolescent girls and young children in the majority of countries^{3,11}.

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OS (-)	Iron source	Solubility	RBA (%)	Comments
0	Iron powders	Very low soluble	75	Unknown corrosion
0	Iron ingots	Very low soluble	n.a	Unknown corrosion
II	Fe(II) sulfate	Highly soluble	100	Least expensive
II	Fe(II) chloride	Highly soluble	50	Not stable in air
III	Fe(III) ammonium citrate	soluble	51	Soluble, not bioavailable
III	Fe(III) pyrophosphate	Very low soluble	43	Unknown dissolution, not bioavailable

Table 1. Characteristics of the three main sources of iron for controlling iron deficiency anemia. The most readily available iron sources are selected with two per state of oxidation (oxidation state—OS). RBA stands for the relative bioavailability, RBA is related to the solubility in 0.1 N HCl (Harrison et al. (1976) cited by Kumari and Chauhan⁴).

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The major challenge in combating IDA is the control of the iron bioavailability⁷. Iron sources are widely available on the market but not all of them are bioavailable (Table 1). Table 1 shows that the best iron source (bioavailable) is Fe^{II} sulfate with stable Fe^{II}. Fe^{II} chlorides are also highly water soluble, but not stable under physiological conditions. Fe⁰ and Fe^{III} sources are less soluble and their bioavailability is correspondingly low. In other words, soluble Fe should be present as stable Fe^{II} to be absorbed by the human body^{2,7,17,25}. This is the reason why the simultaneous consumption of Fe rich diets and vitamin C (ascorbic acid—AA) has consistently and successfully treated IDA^{2,7,22,24,26}. Ascorbic acid is a strong reducing agent for Fe^{III} and forms very stable complexes with Fe^{II} [Fe^{II}-AA]²⁷⁻²⁹. Fe^{II}-AA complexes are readily absorbed by the human body. Thus, supplying people with Fe^{II}-AA solutions has the potential to decrease IDA^{7,12}.

This communication presents the design of a device to locally generate Fe^{II}-AA solution for drinking water fortification. Granular Fe⁰ is placed in a column and leached with a solution of AA. A calculated amount of the leachate is added to the drinking water tank to yield the daily needed iron concentration. Tools for the implementation of such devices in day-care centers for children and adults are discussed.

The presentation starts with a description of the Fe/AA system ("Background of the Fe^{II}-AA concept" section), followed by a discussion of current uses of Fe⁰ for IDA control ("Current use of Fe⁰ for IDA control" section). "Fe⁰ leaching with ascorbic acid: proof of concept" section presents some experimental data on Fe⁰ leaching with ascorbic acid (e.g. proof of concept), "Designing a Fe^{II}-bearing unit" section prepares the realization of the concept by discussing a Fe^{II}-AA production unit. "IDA and safe drinking water provision: killing two birds with one stone?" section formulates some recommendations for combining IDA control and safe drinking water provision. A short conclusion ("Concluding remarks" section) closes the presentation.

Background of the Fe^{II}-AA concept

The ability to improve iron status in populations largely depends on the understanding of the biochemistry and absorption of Fe in the human body^{2,7,17,28}. There are two types of iron: heme (found in red meat) and non-heme (found in plant-based foods) iron. Iron absorption in the gastrointestinal tract is lower for non-heme sources of iron. The literature contains many contradictory findings regarding parameters pertinent to the effective absorption of Fe by humans^{3,5,17}. It seems established that ferrous salts are better than heme iron in combatting IDA, however, some newer iron formulations have claimed the opposite^{30,31}. Fortunately, it is unequivocally reported that a combination of ascorbic acid (AA) and Fe-bearing diets improves the iron status in populations^{7,30}. Being a weak acid, AA is a strong reducing agent for Fe^{III} and an excellent complexing agent for Fe^{II7,28,29}. This means that, where necessary, AA reduces aqueous Fe^{III} – Fe^{II} and builds the very stable Fe-AA complex which is available for the human body (Fact 1: AA reduces aqueous Fe^{III} and builds stable Fe-AA complex). Therefore, Fe-AA complexes are bio-available in the human body⁷.

People living in a high-iron groundwater setting have demonstrated better iron status or suffer less from IDA³²⁻³⁸. The rationale for this finding is that Fe-rich groundwater contains Fe^{II} which is oxidized to Fe^{III} upon contact with air $(20\% O_2)^{39}$. This implies that the amount of bioavailable Fe^{II} also depends on the duration of storage. Upon oxidation Fe^{III} precipitates as Fe hydroxides/oxides or is complexed to less/non bioavailable species. Fe-rich groundwater contains bioavailable Fe^{II}. Whenever clean Fe-rich groundwater is available as a drinking water source, it suffices to stabilize Fe^{II}, for example by addition of AA or lemon juice, to improve the Fe status of the population^{25,40,41}.

Taken together, supplying populations with drinking water containing Fe stabilized in the ferrous form (Fe^{II}) is sufficient (Fact 1). This idea is not new, as it has been successfully applied in rural Brazil for the last three decades^{7,21–24,26}. In its original form, each family was required to have an earthen pot with about 10 L capacity for storing drinking water. Families received a concentrated iron solution (l0 g/L) in the form of ferrous sulfate (FeSO₄) and L-ascorbic acid in the molar ratio Fe^{II}:AA = 1:3, dispensed in 10 mL bottles²¹. The success of this approach has motivated its extension to day-care institutions^{7,24}. The present work seeks to leach Fe^{II} from metallic iron (Fe⁰). Thus, commercial ferrous sulfate is replaced by a more affordable granular Fe⁰, which is additionally readily available, for example as iron filings, iron wire, scrap iron, sponge iron (direct reduced iron), or steel wool. Fe⁰ sources to be considered in this context should not contain any toxic alloying elements. Sponge iron is certainly the best material fulfilling this prerequisite. The typical mineralogical composition (in %) of sponge iron is^{42,43}: Fe (total): 92–95; Fe⁰: 85–90; C: 1.0–1.5; S: 0.005–0.015; P: 0.02–0.09, SiO₂: 1.0–2.0, and balanced by

the gange. The gange is the residual unreduced oxides, mainly comprising of Al_2O_3 , CaO, FeO, MgO, MnO, and SiO₂. The typical mass density of sponge iron is 1600 kg m⁻³ and its apparent density is 3200 kg m⁻³⁴².

Materials selected herein for use in the Fe fortification unit are known to be effective for producing stable soluble Fe^{II} solutions under environmental conditions. Availability and cost are also considered in the selection process because substantial quantities would be required for decentralized production. Used Fe⁰ particles should not contain toxic alloying elements (e.g. Cr, Ni). Fortunately, this is the case for many readily available Fe⁰ materials such as cast iron and low alloyed steel. For example, Lufingo et al.²⁹ analyzed nine commercial steel wool for their elemental composition and found that the Fe⁰ content was constantly higher than 99%, while the (Cr + Ni) level was lower than 0.7% in all the specimens. These data suggest that a solution containing some 10 mg/L of Fe would contain non-detectable levels of (Cr + Ni). However, ideally, used Fe⁰ specimens should be free of Cr, Ni and Pb. Therefore, there is a need to (1) determine the chemical composition of potential Fe⁰ materials, and (2) test them with regard to the leaching ability of relevant toxic elements before their use for drinking water fortification. The next section presents a proof of concept, limited to illustrating the Fe leaching capability of a 0.02 M AA solution (pH = 3.5) from four selected Fe⁰ specimens, in five parallel experiments.

Current use of Fe⁰ for IDA control

Fe⁰ is currently considered an adventitious source of bioavailable iron with both adverse and beneficial effects on human health (Tables 1, 2)^{44–50}. On the one hand, excessive Fe intake (e.g. Fe overload or iron poisoning) is attributed to metallic poisoning derived from foods and drinks prepared in Fe⁰-based vessels^{2,18–20,44,45}. On the other hand, Fe leached from Fe⁰-based cooking utensils is recommended to prevent and cure IDA^{18–20,40,49,50}. Where Fe⁰ cooking utensils are not available, not affordable, or not socially accepted, reusable Fe⁰ ingots have been used^{51–55}, for example in Al-based cooking utensils (Fig. 1). Figure 2 shows the photograph of a fish-shaped iron ingot as used for in-situ food fortification in Cambodia as well as a leaf-shaped iron ingot as used in India^{55,56}.

Tool	Status	References
Leaching Fe from cooking pots	Field applications	57
Placing Fe ⁰ in drinking water containers	Experimental	38
Leaching Fe from water pipes	Experimental	38
Placing Fe ⁰ ingots in cooking pots	Field applications	58
Leaching Fe from Fe ⁰ particles	Conceptual	This study

Table 2. Overview on the status of current approach to exploit metallic iron (Fe⁰) to controlling iron deficiency anemia (IDA).

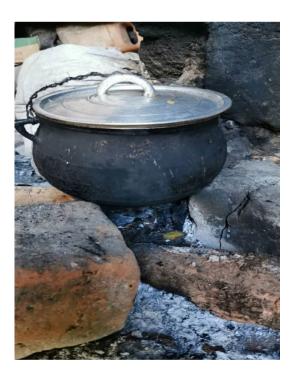


Figure 1. Photograph of an aluminum cooking pot on the fire in Bamena (rural Cameroon). Photograph taken by Serge Ndokou-Nana, October 2021.



Figure 2. Photograph of a Lucky Iron Fish and a Lucky Iron Leaf. Photograph taken by Gerhard Hundertmark, November 2021.

Fe⁰ in the form of iron powders has also been widely used in food fortification^{14,17,56}. In this context, Fe absorption is governed by the extent of the dissolution of used Fe⁰ powders in the gastric fluid¹⁷. The extent of Fe absorption thus depends on the intrinsic reactivity of the used Fe⁰ in the human gastric fluid. The lack of characterization of the Fe⁰ intrinsic reactivity seems to be a major shortcoming as many different Fe⁰ types have been tested and used without appropriate quality control⁵⁸. A proper quality control would characterize the relative Fe bioavailability from used Fe⁰ powders. For example, H-reduced Fe⁰ powders for fortifying cereal flours have been largely used, while the WHO recommends only electrolytic iron powder¹⁴. The WHO recommendation is based on field evidence from Fe absorption in efficacy studies. However, it would have been better to develop an operational parameter (e.g. a dissolution index) to assess the Fe⁰ dissolution trend under different physiological conditions (in the gastrointestinal tract).

While it is clear that Fe⁰ is a relevant Fe source to cure and/or prevent IDA, it is not clear why a specific Fe⁰ source is preferentially used, and which specific operational conditions are optimal to meet the daily needs of a human being^{2,7,17,23}. Relevant Fe⁰ sources include: (1) Fe⁰ cooking and storing vessels (pots and drums)^{44,45,59}, (2) reusable Fe⁰ ingots^{51-53,55,60} (https://luckyironfish.com, Access 2021/10/25), and (3) Fe⁰ powders^{2,56,61,62}.

Food fortification is largely considered the best strategy to increase iron intake of a population, especially for children and pregnant women^{2,14–16,55}. The WHO has recognized food fortification as a potential universal tool for defeating IDA worldwide¹¹. However, there are several concerns to be named: (1) by solving one problem (IDA) in some people, universal fortification exacerbates Fe intoxication for other people (questioned universality), and iron poisoning is as severe as IDA², (2) because of low income, a large fraction of the population has only restricted access to commercial fortified foods (questioned affordability), and (3) it is not known which fraction of Fe⁰ in food is effectively solubilized during digestion and which proportion is absorbed by the body of each individual person^{2,7,17,23}.

With the objective of solving the three named problems this communication suggests a solution that is beneficial to the segment of the population (potentially) suffering from IDA. This solution is called 'semi-universal' fortification² and uses water as a vehicle⁷. Moreover, only drinking water is fortified and is considered affordable or at least more affordable than commercial fortified foods. Concerning the bioavailability, Fe is leached from granular Fe⁰ by ascorbic acid and is long-term stable and bioavailable^{7,21–24,26,28}.

Fe⁰ leaching with ascorbic acid: proof of concept

Fundamental aspects. The present study presents a concept to extract Fe^{II} from Fe^0 specimens, using ascorbic acid (AA) as leaching agent or lixiviant. Previously, AA has been used to leach and extract metals from natural metal oxides (e.g. marine MnO₂) by reductive dissolution^{63–66}. In this context, AA is a reduction and leaching (chelating) agent for ore processing at ambient temperature and under normal pressure. A key lesson from this hydrometallurgical process is that AA leaching has good dynamic characteristics, high reaction kinetics, and requires simple equipment. In this paper, AA is used to sustain the oxidative dissolution of Fe^0 specimens. Fe^0 is oxidized by water (H⁺) (oxidative dissolution) (Eq. 1) and the resulting Fe^{2+} is stabilized by chelation

with AA (Eq. 2). In the absence of AA, Fe^{2+} would have been further oxidized to Fe^{3+} by oxygen present in air (Eq. 3) and precipitated as $Fe(OH)_3$ (Eq. 4)^{58,67-70}. From Eq. (1), a tool to increase the extent of Fe^{2+} leaching is to lower the pH value (H⁺ addition).

$$\mathrm{Fe}^{0} + 2 \mathrm{H}^{+} \Longrightarrow \mathrm{Fe}^{2+} + \mathrm{H}_{2} \tag{1}$$

$$Fe^{2+} + AA \Rightarrow Fe(AA)^{2+}$$
⁽²⁾

$$4 \operatorname{Fe}^{2+} + \operatorname{O}_2 + 2 \operatorname{H}^+ \Longrightarrow 4 \operatorname{Fe}^{3+} + 2 \operatorname{OH}^-$$
(3)

$$Fe^{3+} + 3 OH^{-} \Rightarrow Fe(OH)_{3}$$
 (4)

Once $Fe(AA)^{2+}$ complexes are formed (Eq. 2), they remain stable even when the pH increases to values as high as $8.0^{71,72}$. In particular, Conrad and Schade⁷¹ demonstrated that, adding NaOH to a (FeCl₃ + AA) solution results in a soluble iron chelate, while adding AA to a (FeCl₃ + NaOH) mixture results in an insoluble Fe(OH)₃.

Organic acids (e.g. acetic acid, citric acid, oxalic acid) and other chelating agents (e.g. ethylenediaminetetraacetic acid—EDTA) can be used as effective lixiviants for fly ash and minerals^{73,74}. Organic acid mixtures are currently tested to recover valuable metals from spent Li-batteries⁷⁵. For example, the process described by Chen et al.⁷⁴, used iminodiacetic acid and maleic acid to quantitatively recover Li⁺ and Co³⁺ at 60 °C. AA then converts $Co^{3+}-Co^{2+}$ and enables selective recovery of Co. The present work uses AA to sustain Fe⁰ dissolution (Eq. 1). Comparable approaches are efforts from our research group using two organic chelates (EDTA and 1,10-Phenanthrolin) to characterize the intrinsic reactivity of Fe⁰ specimens^{29,58,76}. Moreover, our research group has been routinely using a 0.1 M AA as a washing solution to free glassware from Fe^{III} oxides after Fe⁰ decontamination experiments.

Experimental procedure. This section is adapted from Ndé-Tchoupé et al.⁷⁶ who characterized the reactivity of twelve Fe^0 materials for H_2 evolution in H_2SO_4 . The four tested herein were included, and depicted significant different reactivity. This result was recently confirmed using a newly developed test for Fe^0 screening: the ascorbic acid test⁵⁸.

Solutions. The working solution was prepared from a L-ascorbic acid powder (Merk, Darmstadt, Germany). The used 1,10-Phenanthroline, sodium ascorbate, and the iron standard (1000 mgL^{-1}) were also from Merck (Darmstadt, Germany). All chemicals were of analytical grade.

Iron materials. Four selected Fe⁰ materials were used. Two of them were commercially available materials for groundwater remediation termed as: (1) "sponge iron", and (2) "iPuTec". Sponge iron is Eisenschwamm from ISPAT GmbH, Hamburg; while iPuTec is Graugußeisengranulat from iPutec GmbH & Co. KG, Rheinfelden; both in Germany. The other two materials were scrap iron materials from a metal recycling company (Metal-laufbereitung Zwickau) termed as: "S15" and "S69". S15 was a mixture of mild steels from various origins, while S69 was a similar mixture of cast irons. Apart from sponge iron, Fe⁰ materials were used in their typical state and form (i.e., "as received" state). Sponge iron was crushed into small pieces, sieved and the particles with sizes ranging between 1.0 and 1.6 mm were used, without any further pre-treatment.

Table 2 summarizes the elemental compositions of the materials based on analyses made using X-Ray fluorescence spectrometry. It can be clearly seen that the materials primarily differ in their carbon (C) and silicon (Si) contents. Thus, based on the C content, the tested materials can be divided into three classes: (1) iPuTec and S69 containing more than 3% C (cast irons), (2) S15 containing less than 2% C (mild steel), and (3) sponge iron (1.9% C), belonging to the third class, characterized by a specific manufacturing technology, which yielded porous materials^{42,43}. All these materials were irregular in shape (filings and shavings) with rough surfaces. Sponge iron had a very rough surface and was even porous. iPutec and the two scrap irons (S15 and S69) were visibly covered with rust.

Experimental methods. 1.0 g of each Fe⁰ material was placed in a chromatographic column containing sand in its lower third and the 0.02 M AA solution in its upper two thirds (Fig. 3). Fe⁰ was leached daily for five consecutive days (Monday–Friday) every week with about 180 mL of a 0.02 M ascorbic acid solution (pH=3.5), at constant temperature of 23 ± 2 °C. At each leaching event, the exact volume of the leachate was monitored and its iron concentration was determined. The experiment was ended after 55 leaching events. This corresponds to a leaching rate of 53% for sponge iron (the most reactive material). An accompanying experiment with 2.0 g of iPuTec was performed to enable the assessment of the impact of the Fe⁰ mass on the extent of Fe leaching by AA.

Analytical methods. Analysis for iron was performed using the Phenanthroline method. Although Fe(AA) was already Fe(II), reduction was performed to follow the analytical protocol which include calibrating the standard solutions. Iron concentrations were determined by a Cary 50 UV–Visible Spectrophotometer (Cary instruments, LabMakelaar Benelux B.V., Zevenhuizen, The Netherlands) at a wavelength of 510.0 nm using 1.0 cm glass cells. The pH values were measured by combined glass electrodes (WTW Co., Weinheim, Germany).



Figure 3. Column experimental set-up for Fe^0 leaching by ascorbic acid (2 mM). The photograph was made at the end of the experiments. The spout of the third column was broken during the experiments but this has no incidence on the performance of the system.

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Results and discussion. Table 4 and Fig. 4 summarize the results of Fe extraction from the four tested Fe⁰ materials. It is seen from Table 3 that sponge iron exhibited the highest extent of Fe leaching with 529.5 mg or 53% of the initial 1.0 g after 55 leaching events over 129 days. The increasing order of Fe⁰ reactivity with respect to the extent of Fe leaching in 0.02 M AA is: S15 < iPuTec < S69 < sponge iron. The high reactivity of sponge iron is attributed to its higher porosity and the corresponding surface area in comparison to other materials. The same order of reactivity was reported in related works^{58,76}. Another important feature from Table 4 is the fact that using twice the amount of Fe⁰ (2.0 g for iPuTec) did not double the extent of Fe leaching. In fact, when doubling the initial Fe⁰ mass, the daily leached mass of Fe increased by only 24.4%, from 8.6 to 11.4%. This observation is consistent with the non-linear kinetics of Fe⁰ dissolution⁵⁷.

Figure 4a shows that the daily dose of 2–12 mg of Fe could be leached from each column containing 1 g of Fe⁰. For each material, the leached amount was high at the start of the experiment, then it decreased progressively with increasing leaching events (elapsed time) until about 70 days. It then increased again to values comparable to initial values for all Fe⁰ specimens except S15 until day 110 (Table 5). After day 110, a new decrease of the leached Fe level started. The trend was the same for all Fe⁰ specimens including S15, with only differences in magnitude. Interestingly, around day 70, sponge iron exhibited the lowest extent of Fe leaching. Figure 4b depicts the cumulative extent of Fe leaching and shows clearly that sponge iron is the best material over the 129 leaching events.

A combination of (1) non-constant kinetics of iron corrosion for individual materials, and (2) different laws of the variation kinetics amount materials, make any prediction of the leaching extent challenging (Table 5). Table 5 shows that for the first 10 leaching events, the increasing order of reactivity was iPuTec < sponge iron < S15 < S69. After this initial period, S15 was the least reactive material until t = 112 d, corresponding to the 52nd leaching event. Between the 10th leaching event and the 52nd there is also no uniform trend in the variation of the extent of Fe leaching from the three other materials. However, it is certain that various amounts of Fe(AA)²⁺ can be obtained to prepare diluted solutions to prevent or combat IDA by varying the following factors: (1) the Fe⁰ mass (e.g. 1.0 g, 2.0 g), (2) the Fe⁰ type (e.g. sponge iron, iPuTec), (3) the AA concentration (e.g. 0.02 M, 0.2 M), and eventually (4) acidifying the solution. Fe⁰ can first be leached by EDTA and the resulting solution (Fe^{III}EDTA) reduced and stabilized to Fe(AA)²⁺. In fact, preliminary experiments (results not shown) have demonstrated that EDTA is a far better lixiviant than AA. The ability of AA to reduce Fe^{III}EDTA is documented and used in analytical chemistry^{29,58}.

This experiment has unequivocally shown that using two columns containing the same amount of a Fe^0 specimen (m) will yield a larger leaching Fe level than a single column containing 2 times the same materials (2 * m). This is due to extreme complexity of the phenomena associated with aqueous iron corrosion (Table 1)^{29,57}. Summarized, these results prove that Fe⁰ leaching using AA is a promising approach to generate stable Fe^{II} solutions to improve the iron status of humans.

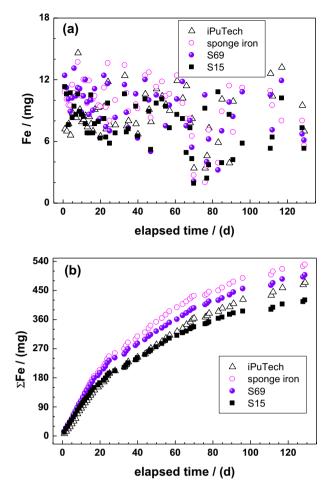


Figure 4. Time-dependent extent of Fe leaching from the four tested Fe⁰ specimens: (a) mass per leaching event, and (b) cumulative masses. Experimental conditions: $m_{iron} = 1.0 \text{ g}$, [AA] = 0.02 M, and T = 23 ± 2 °C.

	Element (%)								
Fe ⁰	С	Si	Mn	Р	S	Cr	Мо	Ni	Fe
Sponge iron	1.96	0.12	0.09	0.027	0.14	0.003	n.d	< 0.001	bal.
iPuTec	3.39	0.41	1.10	n.d	0.105	0.34	n.d	0.088	bal.
S69	3.52	2.12	0.93	n.d	n.d	0.66	n.d	n.d	bal.
S15	0.45	0.28	0.78	n.d	n.d	2.67	n.d	1.34	bal.

Table 3. Elemental composition of iron materials used in this study. *n.d. = not determined and (**)bal. = balance.

Leaching rate		S15 iPuTec S69		sponge iron	iPuTec (2)	
Daily	(mg)	7.6	8.6	9.0	9.6	11.4
Total	(mg)	420	475	497	530	628
Total	(%)	42.0	47.5	49.7	53.0	31.4

Table 4. Summary of the extent of Fe leaching from the tested Fe^0 specimens after 55 leaching events. The daily leaching is the sum of the leaching mass divided by 55, 'iPuTec (2)' corresponds to the experiment using 2.0 g of Fe^0 .

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Event (-)	Time (d)	IPuTec (mg)	Sponge iron (mg)	S15 (mg)	S69 (mg)
2	2	7.0	10.1	10.6	11.2
10	10	7.9	10.4	8.6	8.8
20	22	9.4	11.2	8.2	12.4
30	44	10.6	12.4	10.1	12.0
40	68	7.8	8.2	7.3	8.0
50	96	13.0	11.2	5.8	10.8
51	111	12.6	10.9	5.3	10.5
52	112	10.4	9.5	6.4	7.1

Table 5. Daily Fe leached mass (mg) from 1.0 g of the tested Fe^0 specimens at 8 selected events. The Fe^0 specimens are ordered from left to right in the order of increasing value of m after the second leaching event, corresponding to day 2 of the experiment.

Designing a Fe^{II}-bearing unit

The conceptual design of a Fe^{II}-AA production unit involves two components: (1) a reactive source of metallic iron (Fe⁰), and (2) an ascorbic acid (AA) solution. In principle, batch and column leaching operations are possible. However, column operations are preferred herein mainly because they can run for several months with limited labor input ("Fe0⁰ leaching with ascorbic acid: proof of concept" section). "Fe⁰ leaching with ascorbic acid: proof of concept" section). "Fe⁰ leaching with ascorbic acid: proof of concept" section and available data on Fe⁰ leaching by ethylenediaminetetraacetic acid (EDTA)²⁹ suggest that it is possible to leach constant amounts of Fe from Fe⁰ filling, sponge iron, and steel wool placed in a glass column for several weeks⁷⁷ (Fig. 5). The Fe concentration in the effluent (C₀) depends mainly on the intrinsic reactivity of used Fe⁰, the Fe⁰ mass used, the flow velocity of the AA solution, and the AA concentration. The C₀ value (Eq. 5) is selected such that a certain volume of the effluent (V₀) is added to a water reservoir (V₁) to obtain the desired concentration of Fe in the fortified drinking water (C₁).

$$C_0 = C_1 * V_1 / V_0 \tag{5}$$

Assuming that available safe drinking water is iron free, the mass balance of Fe implies that $C_0V_0 = C_1V_1$ (Eq. 1). If 1 m³ Fe^{II}-AA fortified water ($V_1 = 1000$ L) containing 2 mg/L Fe^{II} (C_1) is to be daily produced, and 1.0 L (V_0) of the effluent should be used, then the C_0 value should be 2000 mg/L or 2.0 g/L ($C_0 = 2000$ mg/L). The challenge is to find the best combination of Fe⁰ materials (e.g. iron filings, sponge iron), Fe⁰ mass, AA concentration,

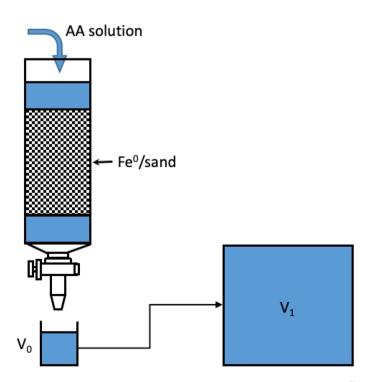


Figure 5. Schematic representation of the process of generating the Fe^{II} -AA solution (V₀) and adding it to a safe drinking water storage tank (V₁). Fe^0 is a reactive iron source. Sand is used as a filling material.

and flow velocity of the AA solution, yielding 1 L of a 2.0 g/L Fe^{II} effluent. In case 2.0 g/L Fe is not realistic, one should rather seek to have 10 L of the effluent with 0.2 g/L Fe^{II} ($C_0 = 200 \text{ mg/L}$).

Figure 5 shows an operational device for the production of the Fe^{II} -AA effluent for dilution. For the realization of this concept, common affordable laboratory devices for weighing (Fe⁰, AA) and analytically determining Fe are needed. This means that for the development of the Fe^{II} -AA method, a small chemical laboratory or Fe^{II} sensor is necessary. However, once the method is established, a laboratory is no longer necessary, and trained personnel can build columns to leach Fe^{0} and perform dilution in water tanks. Calculations are made herein for 1 m³. For larger populations, the 1 m³ water device can be used as a module, and as many modules as necessary can be used to cover the needs. The operational C₁ value of 2 mg/L is purely arbitrarily considered. More relevant values should be selected for testing.

A survey of the literature reveals that various Fe doses have been administered to persons in individual studies. For example, Ginanjar et al.²⁵ discussed the results of some previous studies using oral supplements of a 0 mg (placebo) to 100 mg (therapy) Fe dose in 200 mL water. Fe was added either as $FeSO_4$ or NaFeEDTA and was administered to test persons after at least eight hours of fasting. In other words, up to 100 mg of Fe represents the daily dose to prevent and/or cure IDA. On the other hand, Rakanita et al.¹³ reported that women need 30–60 mg Fe/day. The World Health Organization recommends up to 30 mg Fe/day for children under five^{11,54}. Table 6 summarizes the masses of $FeSO_4$, Fe^{II} fumarate, Fe^{II} gluconate, and NaFeEDTA necessary to obtain 1 kg of elemental iron (Fe). It is seen that (1) 3.0 to 8.0 kg of salts are needed where just 1 kg of Fe^0 suffices, (2) $FeSO_4$ is more than 50 times the price of iron nails (Fe⁰). However, the (bio)availability of Fe from Fe⁰ is primarily uncertain. To design an appropriate Fe^{II}-AA production unit, Eq. 5 is used. The system is operated such that three liters of the drinking water (C₁) bring the needed daily Fe dose for IDA prevention. For curative issues, (up to 100 mg/d), appropriate designs can be developed on the same basis.

Fe leaching as used herein is extensively employed in extractive metallurgy and in reclamation of mining $media^{79-82}$. The operational parameters impacting the effectiveness of the leaching process include concentration of the AA solution, duration of the leaching operation (long-term corrosion rate), Fe^0 grain size, Fe^0 intrinsic reactivity, flow velocity of the AA solution (contact time), and leaching temperature. Given that the kinetics of iron corrosion are neither constant nor linear (see "Fe⁰ leaching with ascorbic acid: proof of concept" Section)^{29,82-84} the service life of each Fe^{II} -AA production unit (Fig. 5) cannot be predetermined. In other words, the question on when to recharge a Fe^{0} /sand column with fresh Fe^{0} can only be answered by testing.

IDA and safe drinking water provision: killing two birds with one stone?

The presentation until here has revealed that many low-income settings are still seeking for reliable ways out of the iron deficiency crisis. Past Fe⁰-based attempts to overcome this problem include: (1) using iron cookwares, (2) adding iron ingots while cooking with aluminum cookwares, and (3) consuming food fortified with Fe⁰ powders. The latter is not suitable because of limited access to commercial fortified foods especially for low-income and vulnerable households. All three tools suffer from the natural time-dependent decrease of the kinetics of iron corrosion (decreased corrosion rate or "reactivity loss")^{29,82-87}. On the other hand, limited access to medical care and other costly iron supplements make other available tools for improving iron status less suitable for generalized use in low-income communities.

During the past three decades, a substantial body of evidence has demonstrated that iron intake from drinking water is a powerful weapon against IDA^{7,31,39}. In this context, Fe^{II} is either naturally available, for example from groundwater^{30,31,34,35}, or artificially added, for example as ferrous sulfate (FeSO₄)²¹. FeSO₄ is reported to be the best water-soluble and cheapest iron salt available (Table 6)⁷. Dutra-de-Oliveira et al.²¹ used 10 mg of FeSO₄ and 100 mg of ascorbic acid (AA) per litre of drinking water. 10 mg of FeSO₄ contains 3.7 mg of Fe, 2.1 mg of S and 4.2 mg of O. This implies that just 3.7 mg Fe is needed for 1 L or some 4.0 g for 1 m³ of water. In other words, 1 kg of Fe⁰ will produce more than 250 m³ of Fe fortified drinking water. The price of 1 kg of Fe⁰ (3.00 Euro)⁷⁸ is far less than that of 1 kg of FeSO₄ (Table 6), and Fe⁰ is readily available, for instance as iron nails or sponge iron^{77,88,89}. The advantage of water as a vehicle for Fe is summarized by Dutra-de-Oliveira et al.⁷ as follows: "Water is consumed daily, everywhere by all ages", including children, pregnant women, and adults of all ages. In other words, Dutra-de-Oliveira et al.^{7,90,91} have already demonstrated the success of iron-fortified drinking water to improve the iron status of low-income populations mainly consuming low iron (Fe^{II}) vegetable diet and daily drinking local water⁹⁰⁻⁹⁶. Consequently, provided local water is of drinking quality, a universal solution to defeat IDA is made more accessible and affordable by using the Fe^{II}-AA method presented herein ("Current

Iron source	Formula	M (g/mol)	x (–)	m _{source} (kg)	Price (Eur/kg)
Elemental iron	Fe	56.0	1.00	1.00	3.00
Fe(II) fumarate	C ₄ H ₂ FeO ₄	170.0	0.33	3.03	95.80
Fe(II) sulfate	FeSO ₄ .7H ₂ O	278.0	0.20	4.96	163.40
NaFeEDTA	C10H12FeN2NaO8	367.0	0.15	6.55	299.00
Fe(II) gluconate	C ₁₂ H ₂₄ FeO ₁₄	448.0	0.12	8.00	102.00

Table 6. Common iron salts used for food and water fortification and their corresponding mass to obtain 1 kg of elemental iron. 'x (–)' is the mass ratio of Fe in the salt. The given kg prices of the salts (chemicals) are from Fisher Scientific (https://www.fishersci.com/—Accesses 26/09/2022). The given Fe⁰ price corresponds to commercial iron nails⁷⁸.

use of Fe⁰ for IDA control" section). AA for Fe⁰ leaching is readily, commercially available. For example, in July 2023, 2.5 kg ascorbic acid (vitamin C), food grade can be purchased from Amazon Germany (www.amazon. de) for just 33 Euro.

The past two decades have witnessed the development of affordable solutions for safe drinking supply⁹⁷⁻¹⁰². From these technologies, one is based on filtration on Fe⁰/sand beds^{97,100,103-107}. In principle, it is possible to design a Fe⁰ filter capable of releasing about 2 mg/L Fe^{II} in the effluent. In such a case, it suffices to add a diluted solution of ascorbic acid to stabilize Fe^{II} and make it available to the human body. Research is needed to achieve the proverbial notion of "killing two birds with one stone": (1) safe drinking water, and (2) iron-fortified water, in a decentralized manner. The problem of clean drinking water supply and IDA co-occur or are juxtaposed in low-income countries¹⁰⁸. This points to the novelty of coupling clean drinking water supply based on Fe⁰ filter systems to the fortification of drinking water to overcome IDA.

Concluding remarks

There are three main approaches to control IDA: (1) supplementation with iron and folic acid tablets, (2) fortification with iron salts, metal iron and dissolved iron, and (3) natural food-based approaches. Efforts for wide implementation of the first two approaches have not really been successful in combating IDA over the past three decades^{14,15,109-112}. The third approach is attractive as it focuses on dietary diversification and enrichment of diets with naturally iron-rich foods, but it is difficult to bring it to scale. Thus, more affordable and applicable tools are still needed.

The Fe^{II}-AA approach is an improved version of a 30-year-old method using commercially available highly soluble Fe^{II} salts⁷. Home iron fortification of water supplies bioavailable iron to rural and urban populations and is optimal for mass supply in schools and other institutions. Systematic research is needed to develop scalable Fe^{II}-AA producing units. Well-designed experiments are needed to determine the practicality of several potential Fe⁰ materials to serve as reliable Fe sources and to combat IDA.

Data availability

All data generated or analyzed during this study are included in this published article and its supplementary information files.

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Conceptualization: J.F.K.-T., S.V. and C.N.; methodology: C.N; writing—original draft: J.F.K.-T. and C.N.; writing—review and editing: S.V. and C.N.; supervision: S.V. and C.N. All authors have read and agreed to the published version of the manuscript.

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