Integration of the oral bioaccessibility of trace elements in the human exposure and risk assessment of contaminated soils

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Abstract
Bioaccessibility and bioavailability are often mentioned as parameters that might greatly improve the accuracy of human health risk assessment of contaminated sites. The objective of this paper is to give some insights into the implemention of using the bioaccessibility concept, focusing on the French methodology. In this framework, the assessment of the exposure media and the more classical human health risk assessment method are the two methodologies defined by the regulatory guidelines. The advantages and limits of integrating the bioaccessibility concept are developed. Bottlenecks are also identified to promote the utilization of such parameters more extensively in the management of contaminated sites.

Introduction
Oral bioaccessibility is defined as the fraction of a soil contaminant that is solubilized in the lumen by the digestive fluids. Oral bioavailability is defined as the fraction that has been absorbed and metabolized. The link between bioavailability (BD) and bioaccessibility (BA) can be formalized as follows (equation 1; Denys et al., 2009) :
BD = BA x AB x M = BA x AB equation

where M is the metabolization of the pollutant (=1 in the case of trace metals) and AB the absorption of the pollutant through the gastro-intestinal membrane.

These two concepts are often mentioned as parameters that might greatly improve the accuracy of human health risk assessments at contaminated sites. As inclusion of this parameter would allow for a more accurate assessment of the exposure, via the oral route. Numerous *in vitro* protocols have been designed to assess the bioaccessibility of soil contaminants. Few *in vivo* protocols have been developed to assess the bioavailability of the contaminants and to validate the physiological accuracy of the bioaccessibility protocols. Recently the Unified Barge [1]Method was validated for As, Cd and Pb and the UBM-bioaccessibility was demonstrated to be linked to the soil-metal speciation (Caboche, 2009). Due to the number of contaminated sites, the amount of land that is affected and the cost and ethic issues linked to in vivo protocols, bioaccessibility appears to be a more useful parameter to include in a site specific risk assessment. However, despite intensive research on the development of *in vitro* methods, no clear guidelines have ever been published related to the possible integration of bioaccessibility data in a quantitative human exposure or risk assessment. The objective of this work is to give some insights into the implementation of bioaccessibility data into the French human health risk assessment methodology. Whereby two regulatory methods might be used: the “assessment of the exposure media” and the more classical quantitative risk assessment. The first method intends to assess the consequence of pollutant emission from a source to its external environment. It consists of a comparison of the concentrations in the different exposure media to either regulatory guidelines or to the local background concentrations.

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Material and Methods
For the “assessment of the exposure media” method, the work was undertaken at the field scale on a former mining site located at Saint-Laurent-le-Minier (Gard). Accordingly to the regulatory guidelines, concentrations of As and Pb in the superficial soil layers (0-5 cm) were measured in the residential areas impacted by the contamination. These measures were compared to the concentrations of As and Pb in the background soils. Then the UBM-bioaccessibility measurements of As and Pb were included in the comparison. For the quantitative risk assessment, the work redefined the initial hypothesis that is implicit in the current exposure assessment calculations for the oral route when the bioavailability or the bioaccessibility of the soil contaminants are not considered. Then, implementation of the bioaccessibility into the dose calculation will be developed. An illustration will be given for As.

Results
In the assessment framework for exposure media, the total concentrations of As and Pb were not statistically significant ($P<0.05$). In this context and according to the regulatory guidelines, it can be defined that there is no difference in terms of human exposure to As and Pb between the contaminated and the background environment. Following this result, no corrective actions to reduce the human exposure in the contaminated environment are needed. However, this result does not consider the potential differences either in speciation or in contaminant distribution within the soil between the impacted and the background environment. Implementing the method including the bioaccessibility of the contaminants allows for

consideration of such differences. In this specific case, and similarly to the total concentrations, the bioaccessibility of As and Pb were not different between the contaminated and the background environment. Thus it confirmed that no corrective action were needed, thereby improving the robustness of this conclusion.
In the framework of the quantitative risk assessment method, the human exposure following soil ingestion is calculated as follows (Equation 2) :

$DI = C \times Qx\ BD_{relative}/P$ equation 2

where DI: Exposure Dose ; C : Soil concentration ; Q : Daily ingested soil ; $BD_{relative}$: Relative bioavailability ; P : Mass

It is currently considered for the exposure assessment that the relative bioavailability of the pollutant between the soil and the reference matrix used to establish the toxicological reference (reference matrix) value is 1. The relative bioavailability is defined as the following ratio (Equation 4)

$BD_{relative} = BD_{soil}/BD_i$ equation 4

where BDsoil: soil bioavailability of the contaminant; BDi: bioavailability of the contaminant from the reference matrix

According to equation 1, equation 4 is equivalent to (equation 5):

$BD_{relative} = (BA \times AB)_{soil} / (BA \times AB)_i$ equation 5

Thus in the framework of the quantitative exposure assessment, the improvement of equation 1 requires knowledge of

- either the bioavailiabilities of the pollutants from both the soil and the reference matrix;
- or the bioaccessibility and the absorption of the substances.

In the case of As, the reference matrix is water. It was recently calculated that bioaccessibility of As was close to 1 and that the absorption of As from contaminated soil is equal to the absorption of As from water (Caboche, 2009). Thus, in this case, equation 5 can be simplified as:

$BDrelative = BA_{sol}$ equation 6

In this case, the measurement of the bioaccessibility of As in soil using the UBM is an easy way to improve the accuracy of the exposure calculation.

Discussion

Integrating the bioaccessibility measured by UBM in the management of contaminated sites allows for improvement of the exposure assessment step. It permits a better integration of the trace elements speciation or distribution within the soil, that are both known as sensitive parameters that influence their bioavailability. It also better reflects the actual human exposure of soil contaminants than the total concentrations, as the UBM was validated against in vivo experiments (Caboche, 2009). However, several bottlenecks do exist before a total integration of the bioaccessibility parameter within the exposure and risk assessment methodologies can be achieved. Firstly, the analytical validation of the UBM should be completed to permit an extensive use of this test by routine laboratories. Second, it is necessary to have more transparency in the construction of the toxicological reference values as precise information on the composition of the reference matrix and data about the bioavailability and the absorption of the element from the reference matrix might be needed.

Conclusion

This paper provides two ways to implement the human exposure and risk assessment regarding the management of contaminated sites. It also allows for the identification of bottlenecks that could be solved by further research programs.

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