

# Role and environmental impacts of copper and zinc in pig farming: from feed to agricultural soils

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■ Zinc and copper are trace metals with a high environmental risk for plants and soil microorganisms if in excess, as well as for animal and human health. There is a risk of accumulation of these elements in soils as a result of repeated application of livestock manure. Due to the low retention of these elements by pigs, most of them are found in the manure. Feeding and treatment of manure are important levers to reduce their excretion and better manage their return to the soil<sup>1</sup>.

## Introduction

Every year, French agriculture and agribusiness produce more than 300 million tons of organic effluents, of which 26 million tons of slurry and 0.8 million tons of manure spread per year in France come from pig farming (Loyon, 2017). These effluents represent considerable sources of nutrients and energy. However, they can also represent sources of air, water and soil pollution at both local and global levels, with harmful consequences on the environment and on animal and human health (OneHealth concept). The challenge is therefore to make better use of the resources present in effluents (nutrients, energy) in order to reduce their environmental impact and to use

their agronomic, energetic and economic potential within the framework of a circular bioeconomy.

Because of their low absorption rate by pigs and their beneficial effects on health and growth performance, zinc (Zn) and copper (Cu) are fed in quantities higher than required. Pig's effluents can then contain quantities of Zn and Cu between 4 and 10 times higher than the needs of the plants when agronomic inputs are reasoned on the basis of nitrogen (N) or phosphorus (P) contents (Jondreville *et al.*, 2002; Revy, 2003). As direct application is the main way of using effluents, these elements are therefore applied in excess and accumulate in soils (Coppenet *et al.*, 1993; L'Herroux *et al.*, 1997; Jensen

*et al.*, 2018), leading to risks of toxicity for soil microorganisms (McGrath *et al.*, 1995) and for plants (McGrath, 1981; Jondreville *et al.*, 2002; Revy, 2003). Furthermore, the use of pharmacological doses (Box 1) of Zn in piglets would contribute to the development of bacteria resistant to antibiotics (Jensen *et al.*, 2018), which is also an important issue for their reduction.

In order to limit these various risks, both for the environment and for animal and human health, European regulations are providing an increasingly strict framework for the quantities of Zn and Cu that can be added to animal feed (EU-2016/1095; EU-2018/1039), with nutrition being the main lever for reducing the flows. There are also

<sup>1</sup> This article is adapted from the paper presented at the Journées de la Recherche Porcine in 2022 (Gourlez *et al.*, 2022).

**Box 1. Glossary of terms.**

**Organic amendment:** fertiliser applied to the soil to improve the physical, chemical and biological properties of the soil (Faure *et al.*, 2018; Dictionnaire d'Agroécologie).

**Bioavailability:** proportion of an ingested nutrient that is absorbed and used by the animal (O'Dell, 1989).

**Pharmacological doses:** For Zn, these doses are 2,500 to 3,000 mg.kg<sup>-1</sup> DM in the feed, for Cu, they are 250 mg.kg<sup>-1</sup> DM.

**Fertiliser:** fertiliser that provides nutrients to meet the needs of plants and soil microorganisms (Faure *et al.*, 2018; Dictionnaire d'Agroécologie)

**Metallothionein:** metalloprotein involved in the detoxification of cells by eliminating free radicals and having a strong affinity for Zn and Cu. This protein has an antioxidant property (Suttle, 2010).

**Supra-nutritional levels:** levels of mineral intake in the feed well above the animal's needs.

**V-scraping:** (see paragraph 2.3.b).

**Speciation:** Speciation is the identification and quantification of different species, types or stages of metals. It considers the chemical and physical properties of elements, i.e. their chemical form, isotopic composition, oxidation state, chemical coordination and molecular structure (Formentini *et al.*, 2016; Kumar *et al.*, 2021). (See section 3.2).

regulations on the amount spread of these minerals, and specifications or standards on the Zn and Cu content of organic fertilisers and amendments. Furthermore, the various effluent treatment technologies that are being developed (composting, aerobic and anaerobic digestion, etc.) can lead to an increase in the concentration of these elements, particularly in relation to dry matter (DM) (Hsu and Lo, 2001; Legros *et al.*, 2017). The knowledge and management of these flows in a context of diversification of effluent recovery procedures are thus important issues.

In this review, Zn and Cu flows and their fate are characterised along the continuum of feed-animal-manure-treatment-soil. The first part refers the use of Zn and Cu in pig feed and the needs of pigs. The release of these elements in effluents and their behaviour in the different pig effluent management systems are described in the second part. Finally, the impact of the return to the soil of products from pig farming and their Zn and Cu composition on the environment are discussed. Due to the limited data available on sows, this review focuses on piglets and fattening pigs.

## 1. Zinc and copper in pig feed

Zn and Cu are considered as essential minerals in pig nutrition. Their bioavailability in the raw materials of the basic feed ration is generally not sufficient to cover the physiological needs (Männer, 2008). Therefore, they are supplemented in the feed in quantities ranging from nutritional levels, in order to cover the animals' needs, to supra-nutritional levels (Box 1) to improve growth or digestive health. However, as their retention is very low, the majority of them are found in the faeces. Reducing the dietary intake of Zn and Cu is therefore the main lever for reducing their quantity in effluents and their potential impact on the environment, while ensuring that they do not affect the animal's growth or health. To do this, it is essential to better understand their function and fate in the body, which influence their bioavailability and therefore their absorption and excretion.

### ■ 1.1 Functions of zinc and copper in pigs

#### a. Body contents

Zn is highly present in the pig's body, with quantities in the order of 1.5 to 2.5 g for an animal of 100 kg live weight (LW). It is found mainly in the muscle

tissue of the pig (about 60%) and in the skeleton (about 30%), with the bones constituting the main site of body reserve, i.e. storage and mobilisation of Zn when intakes vary. The highest concentrations of Zn are found in the bristles (200 mg.kg<sup>-1</sup> DM) and the liver (150 mg.kg<sup>-1</sup> DM). As for most mammals, the plasma Zn content is about 1 mg.L<sup>-1</sup> in pigs, corresponding to 0.1% of total body Zn (Swinkels *et al.*, 1994).

The body content of Cu in pigs is lower, in the order of 200 mg for an animal of 100 kg body weight. Most of the Cu is found in the skeleton (40-46%), muscles (23-26%) and liver (8-10%) (Cromwell, 1997). The liver is the main site of Cu accumulation when requirements are exceeded. Liver Cu content measured after slaughter is used as an indicator of the Cu status of the animals and an indicator of feeding practices. Hodges and Fraser (1983) observed a large variability of Cu content in liver samples from pigs of different origin and age. In 9% of cases, they observed a content lower than 12 mg.kg<sup>-1</sup> DM, indicating a risk of deficiency, but for the majority of animals the content exceeds 20 mg.kg<sup>-1</sup> DM, indicating supra-nutritional Cu intakes. Liver Cu content depends not only on

the dietary concentration but also on the bioavailability of the source (Romeo *et al.*, 2018).

### b. Physiological and catalytic functions

Zn is essential to the proper structural and functional integrity of almost 200 transcription factors. In addition, most metabolic pathways are dependent on one or more proteins that function through the presence of Zn, which is a cofactor in over 300 metalloenzymes (Suttle, 2010). In particular, it is a component of DNA and RNA synthetases and transferases, and many digestive enzymes (National Research Council, 2012).

Cu is an essential element involved in many biological functions, such as cellular respiration, protection against oxidative stress and iron (Fe) transport (Suttle, 2010), as it is present in a large number of enzymes, co-factors and proteins (Espinosa and Stein, 2021).

Zn and Cu play a role in protection against oxidative stress. This is due to their role as cofactors of proteins and enzymes that protect against oxidative stress (superoxide dimutase, ceruloplasmin ferroxidase) (Suttle, 2010). Zn deficiency increases the susceptibility of endothelial cells to this stress, as Zn is the activator of metallothionein (Box 1), which are involved in the detoxification of cells and have a high affinity for Zn and Cu. Moreover, metallothionein also have an antioxidant property (Suttle, 2010).

At “pharmacological” levels, Zn and Cu can also act as “growth promoters” in pigs. This effect may be related to an improvement in feed digestibility or animal appetite (Suttle, 2010), a reduction in digestive disorders, in particular diarrhoea (Bikker *et al.*, 2016), and also to their antibiotic activity and their effect in modulating the

microbiota and improving the integrity of the intestinal barrier (Villagómez-Estrada *et al.*, 2020a)

## ■ 1.2. Pig requirement

The requirement for an element is the amount needed to enable its body to perform all its physiological functions (Revy, 2003; Schlegel, 2010). It can vary according to the response criteria used to define it. As the satisfaction of the physiological requirement of animals depends on the bioavailability of minerals in the feed (National Research Council, 2012), the definition of the requirement must also consider the forms of intake. In practice, it is necessary to know the Zn and Cu requirements and their evolution with the age of the animal, in order to adapt the feed as best as possible and thus reduce the risks of deficits, which are harmful to performance, or excesses, which are harmful to the environment.

### a. Assessment of piglet and growing pig requirement

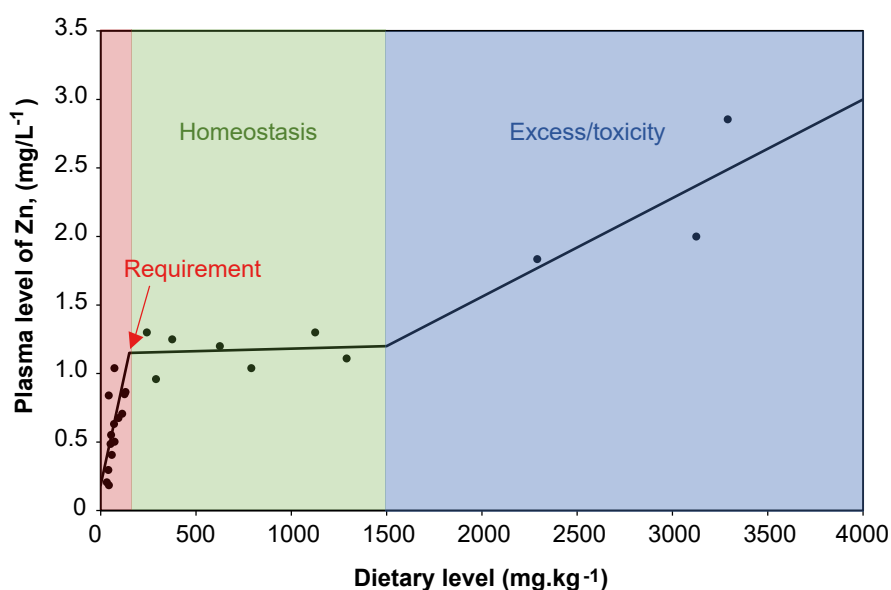
Two main approaches are used to assess mineral requirement, one

empirical and one factorial (Schlegel, 2010), the former being the most common for trace metals (TM). This consists of experimentally testing the effects of a gradual increase in the amounts of Zn and Cu in the feed in order to evaluate the response of several zootechnical or physiological parameters. Statistical treatment of the results by a linear plateau model is then generally used to describe the response and define the breakpoint requirement (Kirchgessner, 1993).

A specific and representative response of one of the animal's functions for Zn or Cu must be observed in order to determine a criterion as characteristic of the requirement (Schlegel, 2010). For Zn, the most representative criterion generally used is its concentration in bone or plasma; for Cu, this criterion is its concentration in the liver (Jongbloed, 2010).

This is illustrated in figure 1 for Zn in post-weaning piglets, based on different literature references. Depending on the level of intake, we can define i) a deficiency zone (in red) in which the intake is not sufficient to maintain an

**Figure 1.** Evolution of plasma Zn concentration in post-weaning piglets as a function of the Zn content of the feed (Hahn and Baker, 1993; Hill *et al.*, 2001; Revy, 2003).



adequate plasma level; ii) a homeostasis zone (in green) in which the plasma level remains constant thanks to the implementation of different homeostasis mechanisms (storage, excretion...) and iii) a zone of excess (in blue) in which the plasma concentration remains constant.) and iii) a zone of excess (blue) in which the plasma concentration increases with food intake, the homeostasis mechanisms being probably saturated. The breakpoint between the red and green zones defines the nutritional requirement, whereas the breakpoint between the green and blue zones indicates the appearance of a risk of excess associated with a drop-in intake.

The National Research Council (2012) gives different references for Zn requirements for pigs depending on the growth or physiological stage of the animal. For weaners the requirement is estimated at 80 mg Zn per kg feed. For fattening pigs, the average requirement estimated in a basic ration by the NRC (2012) is about 50 mg.kg<sup>-1</sup> DM of feed. This requirement may vary according to different criteria that modify the bioavailability of Zn: the nature of the feed, its calcium (Ca) and phytate content, and the incorporation of phytase (Revy, 2003; Spears and Hansen, 2008).

The strict requirement for piglets for Cu is estimated at 5-6 mg.kg<sup>-1</sup> DM of feed and does not exceed this value for later physiological stages (National Research Council, 2012). However, this requirement does not consider the beneficial effects of higher supplementation levels on growth performance or digestive health at certain critical phases of the animal's life, such as weaning, or for breeding sows. As for Zn, the Cu requirement may vary according to different criteria that modify the bioavailability of Cu, such as the addition of more Zn in the feed.

### b. Deficiency effect

The main symptoms of Zn deficiency are initially loss of appetite, the onset of diarrhoea and growth retardation. Anorexia is one of the first signs observed and leads to reduced femur size and strength in deficient piglets (Suttle, 2010). Emergent diarrhoea is due to reduced enterocyte turnover (Revy, 2003). In addition, a decrease in plasma or serum Zn level is observed as well as alkaline phosphatase and albumin (Suttle, 2010; National Research Council, 2012). However, animals respond to Zn deficiency by regulating the various intestinal Zn transporters, but this is not sufficient to cover the premature decline in plasma and serum Zn concentration in deficient piglets (Suttle, 2010). The most prominent symptom of Zn deficiency and the last sign to appear is a hyperkeratinisation of the skin called parakeratosis (Suttle, 2010; National Research Council, 2012).

Cu deficiency in pigs are rarely observed and can occur in weanlings fed about 100 mg.kg<sup>-1</sup> MS Fe, 130 mg.kg<sup>-1</sup> MS Zn and 2 mg.kg<sup>-1</sup> MS Cu (Jondreville *et al.*, 2002). Apparent signs of Cu deficiency are anaemia, limb bowing, the occurrence of spontaneous fractures and heart and vascular problems and depigmentation (National Research Council, 2012). Recent results (Dalto *et al.*, 2021), however, indicate a risk of Cu deficiency developing in the case of Zn intake at pharmacological doses (2500-3000 mg.kg<sup>-1</sup> DM), confirming the reduction in plasma Cu observed by Hill *et al.* (2001) in this situation (see section 1.3.d).

### c. Excess effect

Zn is not very toxic for most mammals. However, an excess of Zn (Figure 1) in the feed leads to a decrease of its absorption, an increase of its storage in the bones and in the enterocytes after

binding to metallothionein and therefore an increase in tissue turnover and endogenous Zn secretions. At pharmacological dosage (Box 1), excess Zn intake can be accompanied by a reduction in feed intake and therefore in growth rate (Hahn and Baker, 1983). An alteration of Cu and Fe metabolism can also happen, with risks of deficiencies if the excess situation is prolonged (Dalto *et al.*, 2021). Moreover, the susceptibility of the animal to excessive Zn also depends on the Ca, Fe, Cu and cadmium content of the feed (Revy, 2003).

Situations of Cu poisoning are rare in pigs (Jondreville *et al.*, 2002). However, a long-term distribution of more than 250 mg.kg<sup>-1</sup> DM of Cu can lead to a toxic effect and the symptoms observed are a reduction in haemoglobin levels and jaundice (National Research Council, 2012). This can lead to a reduction in Fe storage in the liver and therefore anaemia, due to a negative reaction of Cu on Fe, which has limited absorption (Jondreville *et al.*, 2002). The tolerance of pigs to high dietary doses of Cu depends on the supply of Zn in the feed and the interactions of Zn and Cu with metallothionein (Jondreville *et al.*, 2002).

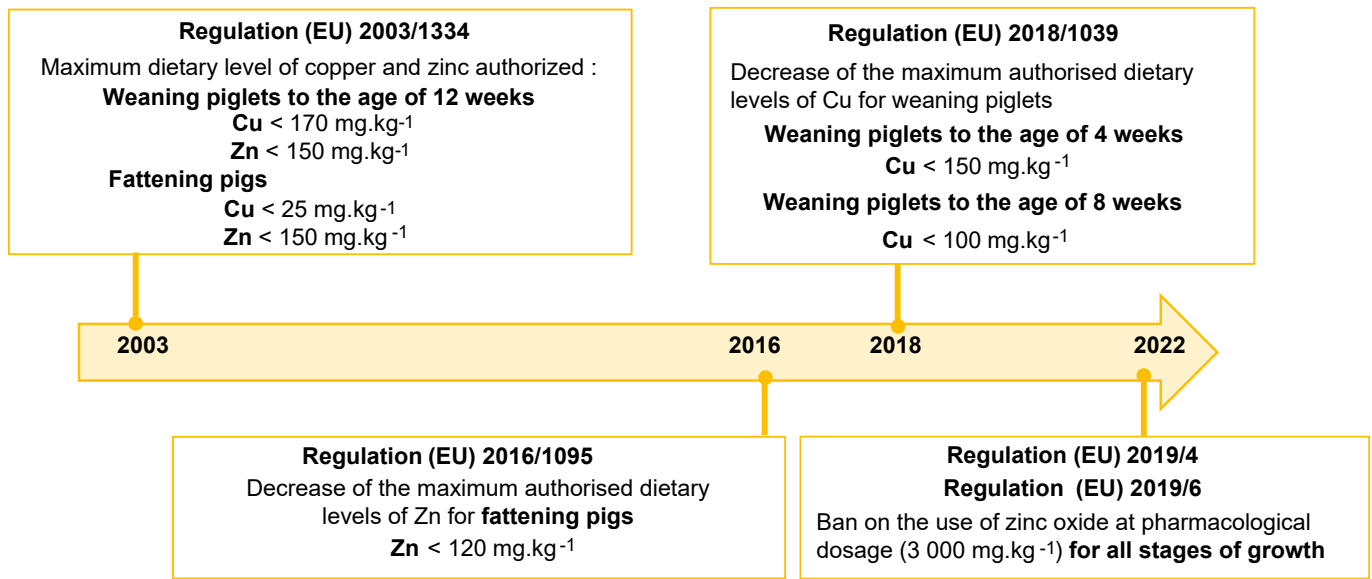
Finally, the capacity of a feed to fulfill the Zn and Cu requirements of pigs and the potential toxic effect of these elements depend on their concentration but also on their bioavailability (Hahn and Baker, 1983; Suttle, 2010); the absorption rate and the corresponding efficiency modify the total TM requirements of the animals (Männer, 2008).

## ■ 1.3. Dietary intake of zinc and copper

### a. Regulation

The European regulations concerning the maximum permitted levels of Zn and Cu in pig feed have changed since 2003 (Figure 2).

**Figure 2.** European regulations on maximum allowed levels of zinc and copper in feed for growing pigs (Regulation (EC) No 1334/2003; Regulation (EU) 2016/1095; Regulation (EU) 2018/1039 of 23 July 2018; Regulation (EU) 2019/4; Regulation (EU) 2019/6).



From 28 January 2022, the use of zinc oxide (ZnO) at pharmacological level was banned in Europe. This regulation aims to reduce the excretion of Zn in faeces (Revy, 2003) and its accumulation in the soil, and to limit the development of antibiotic resistance in animals and humans to which high intakes of Zn contribute (Ciesinski *et al.*, 2018).

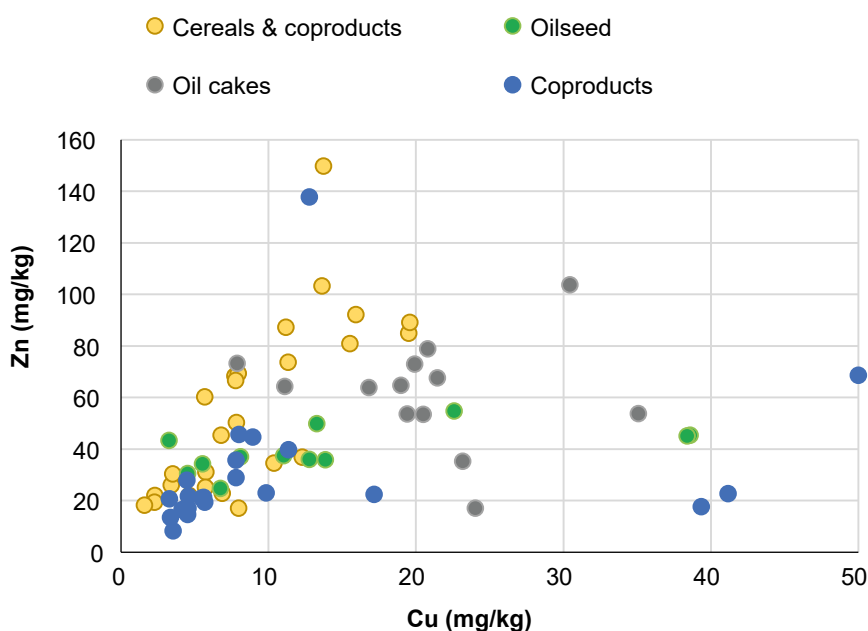
#### b. Composition of raw materials

The basal diet of the pigs is mainly composed of cereals (mainly maize and wheat, but also barley, triticale, etc.), oil cakes (mainly soya, but also rapeseed and sunflower), oilseeds (peas, beans, etc.), cereal co-products (bran, middlings, draff, etc.), and vitamin mineral supplements (feed additives).

Cereals and protein crops contain on average 20-30 mg.kg<sup>-1</sup> DM of Zn, cereal co-products have a higher Zn content (between 70 and 90 mg.kg<sup>-1</sup> DM, Figure 3). Oilseeds and oilcakes contain between 30 and 90 mg.kg<sup>-1</sup> DM of Zn (figure 3).

Cereals, their co-products, oilseeds and oilcakes contain between 5 and 15 mg of Cu per kg DM (Figure 3).

**Figure 3.** Zn content of the raw materials of the basic pig ration as a function of their Cu content (after INRA-AFZ, 2004)



#### c. Different forms of supplementation

Zn supplementation is essential in pig diets in order to cover the animal's needs, as the intake from raw materials alone is not sufficient. Moreover, the variation in the bioavailability of dietary Zn due to the different components of the ration must also be considered (Revy, 2003).

Contrary to Zn, a non-supplemented feed can theoretically cover the Cu requirement of growing pigs (about 6 mg.kg<sup>-1</sup>). However, in practice, Cu supplementation in feed is usually applied to consider the imprecision of the total requirement estimation and to



counterbalance the antagonistic effects of some elements in the ration, which affect the bioavailability of Cu and thus the requirement of the pig. Cu supplementation in the feed is used as a safety margin (Jondreville *et al.*, 2002).

Zn and Cu, when added as additives, can come from different sources. Sulphates ( $\text{ZnSO}_4$  and  $\text{CuSO}_4$ ) are generally considered as the reference sources in comparative bioavailability studies. However, the fact that these sources are highly soluble means that they can also be more easily bound to other elements in the ration, particularly phytates in many feedstuffs, thus reducing their bioavailability (Revy, 2003). There are other sources of supplementation (oxides, chlorides, chelates) which have very variable physicochemical characteristics and bioavailability. In practice,  $\text{ZnO}$  and  $\text{CuSO}_4$  are the most frequently used sources for animal feed (Revy, 2003).

#### d. Bioavailability of copper and zinc from feed and factors of variability

Two experimental approaches exist to measure the bioavailability of a mineral. The first is to measure the response of different physiological criteria to increasing intakes of the mineral, while remaining below the requirement (red area in Figure 1). The second is to measure this response with intakes well above requirement and measure their accumulation in the blood or in different tissues such as bone or liver (Spears and Hansen, 2008). In both cases, these responses are assessed relative to a reference source ( $\text{CuSO}_4$  and  $\text{ZnSO}_4$ ) whose bioavailability is set at 100%. Bioavailability is then calculated by comparing the response slopes of the different sources tested. Generally speaking, the bioavailability of minerals is influenced by the type of mineral

supplementation provided, by factors such as the physiological status of the animal or its mineral status, and also by interaction effects with other agonistic or antagonistic elements of the ration (O'Dell, 1989; Männer, 2008). Different types of possible interactions are the formation of non-absorbable complexes in the gut, competition between cations for the transport of non-specific divalent cations or even the induction of proteins by non-specific metals (Suttle, 2010). The difference in bioavailability of Zn and Cu between sources could be largely explained by these phenomena that affect absorption, as well as their different physicochemical properties that affect their solubility.

Phytates form non-absorbable complexes with Zn, which reduces the bioavailability of Zn (Suttle, 2010). The hydrolysis of phytates allows the release of Zn, but monogastric animals do not naturally produce phytase, or in very limited quantities. The addition of microbial phytase to the ration can therefore improve Zn availability. Revy *et al.* (2004) showed that 700 units of phytase can replace 32-43 mg of Zn in the form of sulphates. Similarly, Bikker *et al.* (2012) estimated that 500 phytase units can replace 27 mg of Zn as sulphate. According to the meta-analysis conducted by Schlegel and Jondreville (2011), only the Zn naturally present in the feed is influenced by phytates and phytase, and not the Zn added to the ration in the form of mineral or organic supplements. This antagonistic effect of phytates is increased after adding Ca to the ration (Suttle, 2010). A phytate-Ca-Zn complex is formed and precipitates Zn in the digestive tract (Revy, 2003). The antagonistic effect of phytates on Zn is all the more important in young animals, which are fed diets providing more Ca than required (Suttle, 2010).

Cu is less affected by phytates and Ca of the ration, due to its higher affinity for free amino acids, with which it forms chelates, allowing Cu to retain its solubility (Jondreville *et al.*, 2002). However, soluble sources of Cu can interact in the digestive tract with phytates and form zinc-calcium-copper-phytate or copper-calcium-phytate complexes (Oberleas, 1973), which are resistant to phytase hydrolytic activity.

Zn has an antagonistic effect on Cu, i.e. it inhibits its absorption. This effect is due to the inducing effect of Zn on the synthesis of metallothionein, which have a strong affinity with Cu, preventing its transfer to the intestinal epithelium (Revy, 2003).

## 2. Zinc and copper excretion by pigs and manure management

Zn and Cu are few retained by animals. They are therefore mainly excreted in excreta, mainly in faeces. The concentrations of these TM can then constitute factors limiting the agronomic use of these excreta. Controlling these levels is therefore important to optimise input strategies according to the effluent utilisation pathway.

### ■ 2.1. Zinc and copper retention and average excreta composition

#### a. Zinc and copper retention in pigs

Several studies have determined the body retention of Zn and Cu by pigs, which makes it possible to calculate the quantities excreted and their content in the effluent by difference with the ingestion. The retention is relatively low and is estimated in the literature for piglets and growing pigs at about  $22 \text{ mg.kg}^{-1}$  of weight gain for Zn (Dourmad *et al.*,

**Table 1.** Zinc and copper discharges from pigs according to CORPEN references (2016), calculated according to RMT Élevage et Environnement (2015) and new estimates taking into account new EU regulations (Regulation (EU) 2016/1095, Regulation (EU) 2018/1039).

Physiological stages	Zn ingested	Zn excreted	Cu ingested	Cu excreted
Breeding sow, g/year	180 <b>144'</b>	173 <b>137'</b>	30.0 <b>30.0'</b>	29.7 <b>29.7'</b>
Post-weaning, g/pig	5.8 <b>5.8'</b>	5.3 <b>5.3'</b>	6.6 <b>4.9'</b>	6.6 <b>4.9'</b>
Post weaning with 3000 mg /kg feed DM, g/pig	19.2	18.7	6.6	6.6
Feeding, g/pig	36 <b>28.8'</b>	34.1 <b>26.9'</b>	6.0 <b>6.0'</b>	5.9 <b>5.9'</b>

<sup>1</sup> According to the new regulations (EU) 2016/1095, (EU) 2018/1039.

2002) and less than 2 mg.kg<sup>-1</sup> of weight gain for Cu (Jondreville *et al.*, 2002). In case of intake close to the requirement, about 80 to 90% of the Zn and Cu ingested by pigs are found in pig faeces of which only 1 to 2% are excreted in urine (Dourmad *et al.*, 2002).

#### b. Average composition of excreta

The composition of the slurry will mainly depend on the composition of the feed given to the pigs. As the digestibility of DM and Organic Matter (OM) of the feed is around 70-80%, and therefore higher than that of Zn and Cu (around 1-2%), these TM are much more concentrated in the manure than in the feed. Manure can thus contain up to 2,000 mg of Zn per kg DM and 1,000 mg of Cu per kg DM, considering all types and stages of animals and feed (Dourmad *et al.*, 2002; Jondreville *et al.*, 2002; Marcato, 2007).

#### c. Reference for copper and zinc release from pigs

In 2016, the official references (CORPEN, 2016) setting the amounts of N, P, Zn and Cu excreted by pigs have been updated (RMT Élevage et Environnement, 2015). They are reported in Table 1, incorporating values calculated considering the most

recent EU regulations on maximum permitted levels in feed (Regulation (EU) 2016/1095, Regulation (EU) 2018/1039).

The levels of Zn and Cu excreted by pigs, calculated considering the new regulations, are lower than the current CORPEN references, which were defined considering the old regulations (Regulation (EC) No 1334/2003). This is due to the reduction of the maximum permitted levels in feed following the new regulations of 2016 and 2018. An update of these references could consider these new values.

### ■ 2.2. Feed strategy and reduction of copper and zinc excretion

There are different dietary levers to reduce Zn and Cu excretion. It is indeed possible to vary either the form of supplementation, or the amount of Zn and Cu added to the feed, or both simultaneously. Using the approach described by Dourmad *et al.* (2013), we calculated the Zn and Cu excretion of pigs between weaning and slaughter (from 8 to 118 kg PV) under different feeding assumptions regarding Zn and Cu levels and considering the evolution of the EU regulations since 2003 and the perspectives for the future. The results presented in figure 4A and 4B clearly

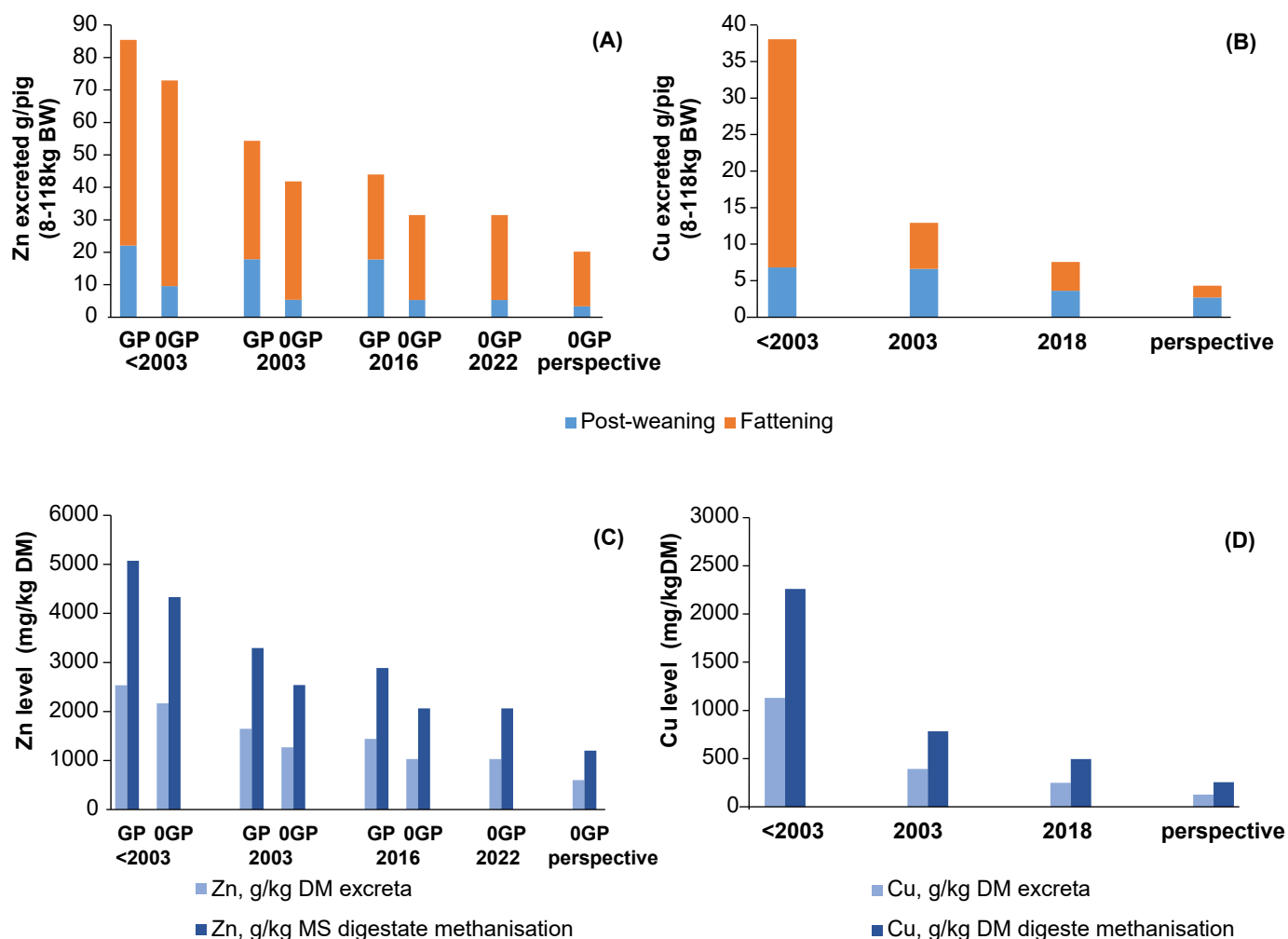
show that the regulatory changes have led to a strong reduction in excretion for both Zn and Cu.

For Zn and with the new regulation from 2022, a 63% reduction in excretion per pig between 8 and 118 kg of PV is observed compared to the situation before 2003 (31.5 vs 85.4 g/pig). For Cu the evolution is even more severe with the current regulation the excretion is reduced by 80% compared to the situation before 2003 (7.6 vs 38.1 g/pig between 8 and 118 kg PV).

This evolution of excretion was also accompanied by a strong reduction in the content of the excreta. For Zn, this content has fallen from 2,100 to 1,000 mg.kg<sup>-1</sup> DM and for Cu from 1,100 to 250 mg.kg<sup>-1</sup> DM. Further reduction prospects can be planned (Figure 4.C and 4.D), but they require further knowledge of animal needs and bioavailability of sources (Dourmad *et al.*, 2013).

The composition of manure also depends on the physiological stages of the animals (figure 5). The Zn and Cu contents relative to DM, depend both on their content in the feed and on the composition of the feed, in particular its fibre content which influences OM

**Figure 4.** Influence of changes in regulations on zinc (Zn) and copper (Cu) content in feed on excretion and effluent content: amount of Zn (A) and Cu (B) excreted per pig between 8 and 118 kg live weight (LW), and average Zn (C) and Cu (D) content of manure and digestate from anaerobic digestion; GP, 0GP: with or without Zn as a growth promoter



Perspectives are 100 ppm Zn for post-weaning piglets and 80 ppm Zn for the fattening pig, and 100 ppm Cu for the first post-weaning period, 80 ppm Cu for the end of post-weaning and 15 ppm Cu in the feed for fattening pigs.

digestibility. Many studies have investigated the effect of modifying the Zn and/or Cu content of the feed on their concentrations in the manure. After reduction of the levels of these TM in the feed (to about 70 mg.kg<sup>-1</sup> DM of Zn and 10 mg.kg<sup>-1</sup> DM of Cu, compared to levels complying with European regulations, i.e. 150 and 25 mg.kg<sup>-1</sup> DM respectively), no effect on the performance of fattening pigs or on carcass quality was observed (Paboeuf *et al.*, 2001; Creech *et al.*, 2004; Van Heugten *et al.*, 2004; Liu *et al.*, 2016; Villagómez-Estrada *et al.*, 2020b). All these studies show that the reduced Zn and Cu feed are effective in reducing Zn and Cu concentrations in manure. Different mineral sources have

also been tested to decrease Zn and Cu excretion (Figure 5).

### ■ 2.3 Pig manure management and impact on zinc and copper

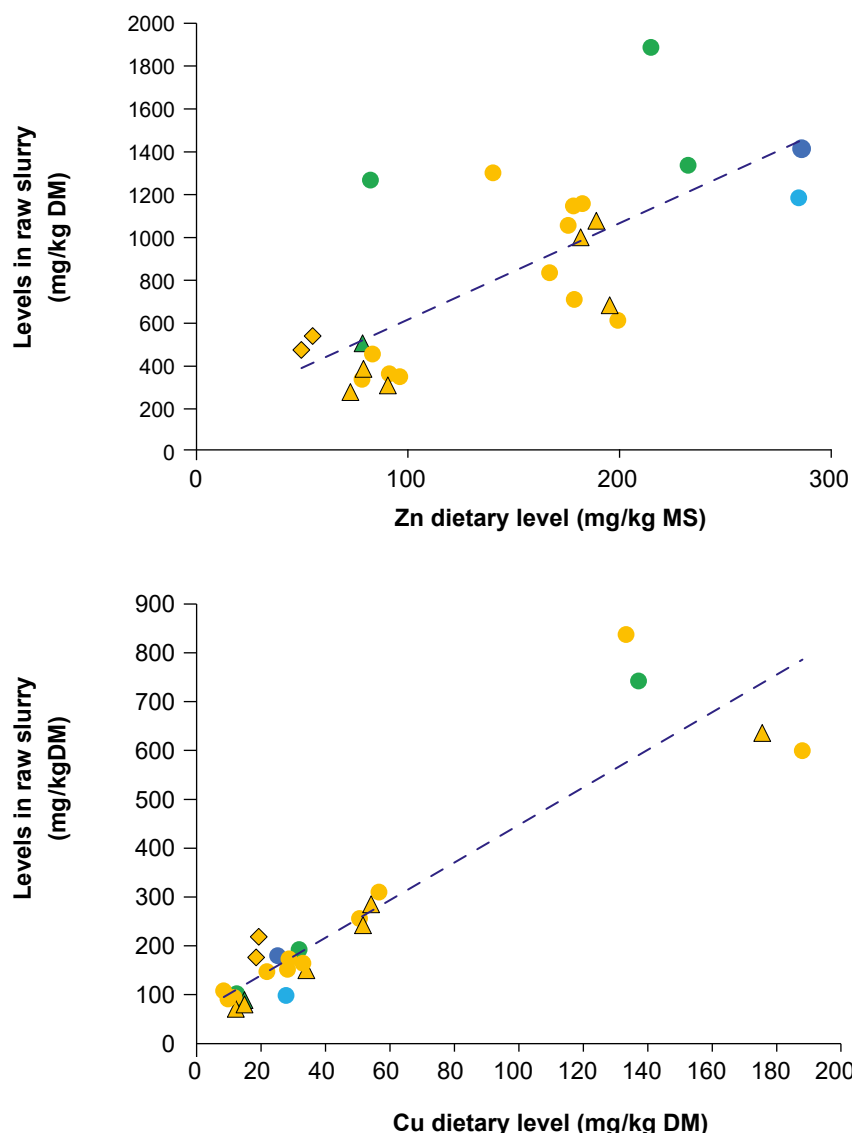
Manure management is an opportunity to better control the distribution of TM in pig manure before it goes back to the soil and thus reduce its environmental impact. Effluent treatment does not eliminate metals but influences their concentration, due to OM degradation and/or phase separations, and their speciation (Box 1), due to different chemical and biochemical mechanisms (adsorption, chemisorption, precipitation...) (Couturier, 2002).

#### a. Pig manure management systems

Although the most common way of managing manure is storage followed by spreading (Nicholson and Chambers, 2008), almost 12% of French pig farms were already using an alternative or complementary manure treatment technique to 'storage-spreading' in 2008 (Loyon, 2017). The treatments applied can be grouped into different categories: mechanical (phase separation), biological (aerobic treatment or anaerobic digestion, composting), chemical and thermal. These different technologies can be combined in different ways and are part of the overall manure management system (Figure 6 and Table 2).



**Figure 5.** Zn and Cu content of manure (mg.kg<sup>-1</sup> DM) as a function of the Zn or Cu content of the feed and the sources of input (after Levasseur and Texier, 2001; Creech et al., 2004; Van Heugten et al., 2004; Hernández et al., 2008; Liu et al., 2016).



Dark blue: pregnant sows; light blue: lactating sows; green: post-weaning piglets; yellow: fattening pigs. Diamond: no supplementation; Round: inorganic source of Zn or Cu; Triangle: organic source of Zn or Cu.

### b. Effect of treatments on zinc and copper in effluents

Table 3 summarises the main effects observed on Zn and Cu during the treatments.

#### • Phase separation in buildings

Phase separation directly in the building, e.g. by the V-scraping process, is fairly recent and the characterisation of the products obtained is still few reviewed. Zn and Cu follow mostly the solid fraction

(between 90 and 95%) and this fraction is mainly treated by composting or anaerobic digestion. (Loussouarn *et al.*, 2014; Likiliki *et al.*, 2020).

#### • Duration and type of storage

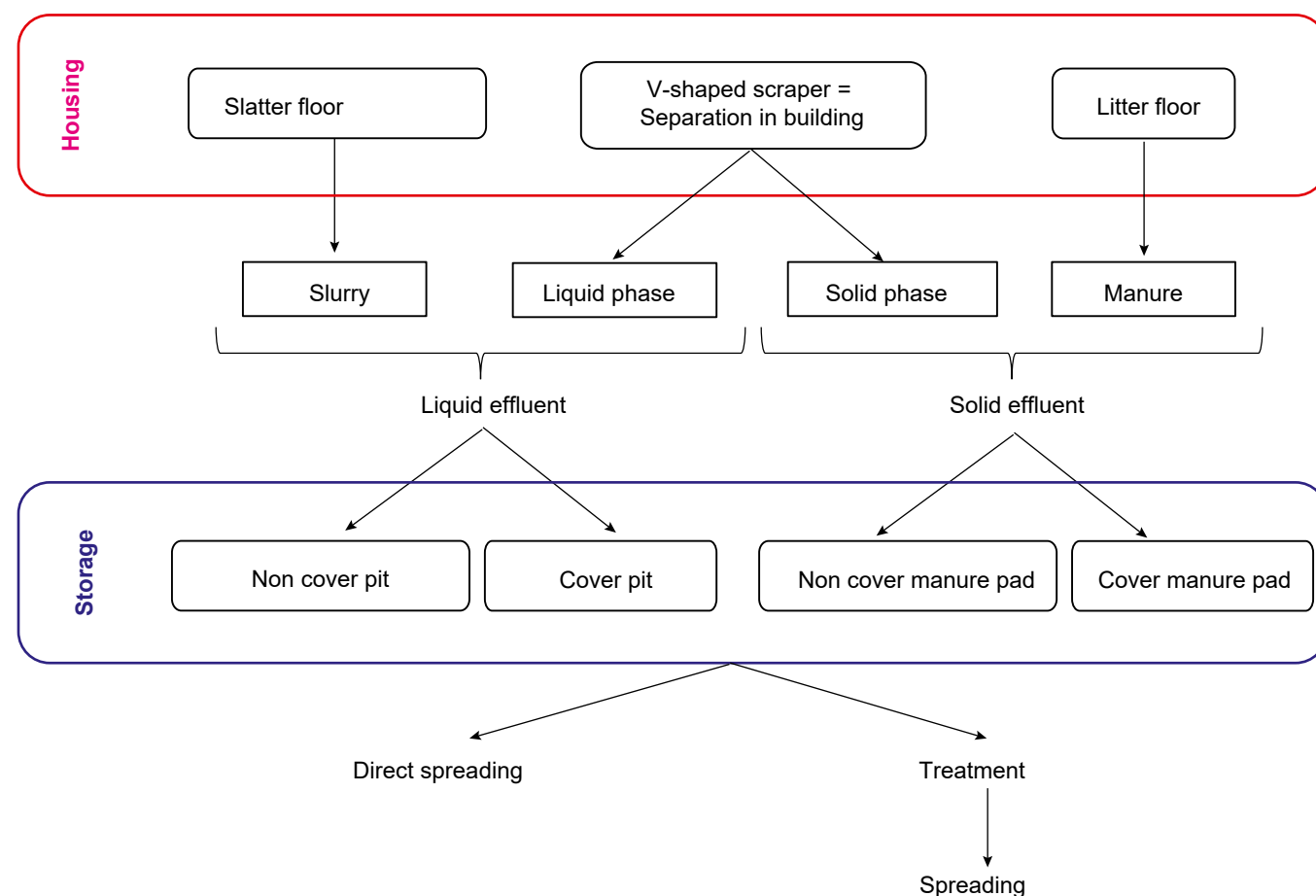
Few studies exist on the effect of storage on Zn and Cu. According to Popovic and Jensen (2012), storage has no significant effect on Zn and Cu concentrations but increases the binding of Zn and Cu to less soluble solid particles.

#### • Mechanical phase separation

As only about 10% of Zn and Cu are soluble, these elements are mainly found in the solid fraction after separation from the slurry (Nicholson and Chambers, 2008). This redistribution will however depend on the type of separator or combination of separation processes used (Møller *et al.*, 2007; Pantelopoulos and Aronsson, 2020). The separation of Zn and Cu to the solid phase is highest for chemical treatment with filtration (99.5% Zn and 94.9% Cu in the solid phase) followed by centrifugation (38.1% Zn and 31.1% Cu) and then screw compacting (7.1% Zn and 6.6% Cu) (Møller *et al.*, 2007; Popovic *et al.*, 2012; Pantelopoulos and Aronsson, 2020). Several studies evaluate the effect of combining different types of separation with an aerobic biological treatment plant (Levasseur, 2003; Beline *et al.*, 2004). The separation results in lower concentrations in the liquid phase, thus reducing the annual soil load of Zn and Cu after the liquid fraction is used as fertiliser by spreading, compared to spreading raw manure (Beline *et al.*, 2004; Møller *et al.*, 2007; Popovic *et al.*, 2012). The solid fraction can then be exported and used as organic fertiliser, usually after further treatment (drying or composting).

#### • Methanisation

As several nutrients such as N, P, K, Zn, Cu are retained during anaerobic digestion, the resulting digestate can be used as fertiliser (Marcato, 2007; Marcato *et al.*, 2009; Amaral *et al.*, 2014). However, the transformation of part of the OM into biogas leads to an increase in Zn and Cu contents in relation to the DM and an effect on their speciation (Box 1) which is important to consider. Furthermore, Zn and Cu can also stimulate or inhibit the bacteria responsible for anaerobic digestion (Matheri

**Figure 6.** Livestock manure and its possible fate (after Møller et al., 2007 Nicholson and Chambers, 2008; Loyon, 2017).

et al., 2016), thus modifying biogas production.

#### • Drying by incineration

After incineration of the solid fraction of slurry or digestate from anaerobic digestion, two types of ash are obtained, a bottom ash (which tends to be richer and to accumulate Cu) and a so-called “fly ash” (Møller et al., 2007).

#### • Composting

Composting leads to the production of humic acid, the main constituent of humus, and Cu has a better affinity for this humic acid than Zn, which influences their distribution (He et al., 2009) in the final product of composting: compost. Hsu and Lo (2001) observed an increase in the soluble Cu content during the first 18 days of the composting process (up to 16% of Cu in

this form) and then a decrease in this content to 3% of Cu in soluble form in the final compost. A lower effect was observed on the soluble part of Zn, which reached a maximum of 2%. Composting therefore seems to have a significant effect on the risk of Cu leaching, but less on that of Zn. Hsu and Lo (2001) observed that most of the Zn and Cu were in the low bio-available fraction of the compost. The addition of material, such as straw or other organic substrates, or mineral additives, reduces the availability of heavy metals during composting and facilitates their transfer to more stable forms. This is related to the formation of compounds with affinity to the humic matter of the compost (Santos et al., 2018; Li et al., 2019). Furthermore, the addition of OM is also accompanied by a reduction in Zn and Cu content through dilution.

One of the purposes of these different treatments is the production of organic fertilisers or soil amendments (Box 1) that are exported outside the livestock farms to crop systems where OM is lacking, such as market gardening. This reduces the risk of accumulation of elements such as Zn and Cu in pig farm soils by optimising their use. However, this requires that these products be standardised and consider the impact of these treatments on the speciation of the TM and therefore their bioavailability to plants.

### 3. Pig manure application and consequences on soils

#### ■ 3.1. Zn and Cu input to soils and regulations

In France, 78% of the Zn and 50% of the Cu brought to the surface of

**Table 2.** Main manure management pathways for pigs (after Møller et al., 2007; Nicholson and Chambers, 2008; Loyon, 2017).

Type of treatment	Type of effluent treated	Products obtained	Next possible treatment
<b>Mechanical Treatments</b>			
Phase separation <i>Sedimentation, screw compactor, centrifugation</i>	Slurry Liquid phase from a separation in a building Digestate from methanisation Purified effluent	Solid phase	Anaerobic digestion Composting Thermal treatment
		Liquid phase	Aerobic treatment
<b>Biological treatments</b>			
Aerobic treatment	Slurry Liquid phase from a separation in a building Liquid phase from phase separation	Purified effluent	Phase separation
Anaerobic digestion <i>Mesophilic (35–40°C), thermophilic (55°C)</i>	Slurry Solid phase from a building separation Solid phase from phase separation Manure	Digestate	Phase separation Heat treatment
Composting	Solid phase from building separation Solid phase from phase separation Manure	Compost	
<b>Thermal treatment</b> <i>Incineration, gasification, thermal gasification, drying, drying, evapo-concentration</i>	Solid phase from a building separation Solid phase from phase separation Manure	Bottom ash Fly ash	
<b>Chemical treatments</b>	Slurry		

agricultural soils come from the spreading of livestock manure (Belon *et al.*, 2012).

#### a. Several sources of input

The Zn and Cu present in soils may come from natural or exogenous sources (e.g. fertiliser inputs). Indeed, metals are found naturally in soils after alteration of the bedrock (Legros, 2008; Kumar *et al.*, 2021). The composition of Zn and Cu and their behaviour depend on the type of soil and the bedrock on which it develops (Legros, 2008). Exogenous inputs are also sources of soil enrichment in Zn and Cu: phosphate fertilisers, inputs of manure and slurry, spreading of sewage sludge, 'urban' composts and phytosanitary

treatments (Baize, 1997). It is therefore important to control Zn and Cu inputs, which can become toxic for plants or soil micro-organisms.

#### b. Regulations on the spreading of livestock manure

Because of the risks associated with the input of Zn and Cu through the application of organic fertilisers and soil amendments, maximum levels of these elements have been set for their commercialisation (Table 4). However, these standards do not apply to raw livestock manure spread as part of a land application plan.

Pig manure for fattening contains about 955 mg.kg<sup>-1</sup> DM of Zn and

300 mg.kg<sup>-1</sup> DM of Cu when this type of animal is fed a feed that complies with current European regulations (120 mg.kg<sup>-1</sup> Zn and 25 mg.kg<sup>-1</sup> Cu), these values being slightly higher if the entire post-weaning/fattening period is considered (figure 5). However, effluent treatment will generally concentrate these elements to a high degree and thus increase their content. Thus, given the current Zn and Cu levels in feed, these levels exceed Afnor standards and even more the European Ecolabel for products resulting from certain treatments. A specific rule was recently established for digestates from methanisation with higher thresholds set at 1,500 and 600 mg.kg<sup>-1</sup> of DM respectively for Zn and Cu (NF U44-051).

**Table 3.** Effects of different pig manure management and treatment on Zn and Cu elements.

Management/ Processing	Treatment effects on Zn and Cu	Parameter to be taken into account	References
Storage	- Increase in the influence of the distribution of Zn and Cu between the different classes of particles: the binding of metals to the particles	Type of storage Duration Temperature	Popovic and Jensen (2012)
Phase separation	- Concentration of Zn and Cu in the solid phase as a function of separation efficiency	Scraping method in the building	Loussouarn <i>et al.</i> (2014) Likiliki <i>et al.</i> (2020)
		Type of separator	Møller <i>et al.</i> (2007) Nicholson and Chambers (2008) Pantelopoulou and Aronsson (2020)
Aerobic treatment	- Increased concentration of Zn and Cu, relative to OM and DM - Redistribution of Zn and Cu between solid (screenings) and liquid phases (supernatant and sludge)	Type of separators	Levasseur (2003) Béline <i>et al.</i> (2004)
Methanisation / Anaerobic digestion	- Increase in Zn and Cu concentration, relative to OM and DM - Increase in the share of Zn and Cu in solid particles - Increased concentration of Zn and Cu of Zn and Cu in the poorly bioavailable fraction available fraction - Decrease in the phytoavailability of these elements	Temperature pH Retention time hydraulic Oxidation-reduction reduction	Marcato (2007) Marcato <i>et al.</i> (2009) Amaral <i>et al.</i> (2014) Matheri <i>et al.</i> (2016) Legros <i>et al.</i> (2017) Yang <i>et al.</i> (2020)
Thermal treatment (incineration)	- Cu accumulation in bottom ash - Concentration of Zn and Cu in particles > 30 µm		Møller <i>et al.</i> (2007) Kuligowski <i>et al.</i> (2008)
Composting	- Increased concentration of Zn and Cu, relative to OM and DM - Increase of soluble part at the beginning of the process and then - Decrease of Cu in this form at the end of the composting process - Decrease in bioavailability factor of Zn and Cu	Moisture pH Nitrogen content Dissolved organic carbon organic carbon content Humic matter content	Hsu and Lo (2001) He <i>et al.</i> (2009) Santos <i>et al.</i> (2018) Li <i>et al.</i> (2019)

**Table 4.** Maximum permitted contents (mg.kg<sup>-1</sup> DM) in fertilisers according to different regulations or standards.

Type of fertilizer	Zinc	Copper
<b>Organic fertilizer</b>		
Ecolabel <sup>1</sup>	300	100
Afnor <sup>2</sup>	600	300
<b>Methanation digestate<sup>3</sup></b>	1,500	600

<sup>1</sup> Decision (EU) 2015/2099.<sup>2</sup> AFNOR standard NF U44-051 (April 2006).<sup>3</sup> Order of 13 June 2017 (NF U44-051 of April 2006).

However, even with these different regulations, the quantities of Zn and Cu applied remain higher than the needs of the crops.

### ■ 3.2 Forms and reactivities of Zn and Cu in effluents

Effluents are generally recovered by spreading on soils and the determination of the speciation of Zn and Cu (Box 1) in these effluents is necessary to better predict the mobility and availability of these elements in soils (Legros *et al.*, 2017). Several analytical methods exist to determine the speciation of elements in a matrix: X-ray absorption spectroscopy (XAS), for example, provides information on the atomic environment around an atom and its degree of oxidation (Legros, 2008). Extraction methods (selective extraction, sequential extraction, etc.) also exist, in order to isolate several fractions with a higher or lower bioavailability and mobility in soils (He *et al.*, 2009).

### ■ 3.3 Impacts of exogenous Zn and Cu input

Zn and Cu are essential trace elements for plant growth, but they are also heavy metals with a high environmental pollution potential (Kickinger *et al.*, 2009). In high concentrations in effluents, Zn and Cu can accumulate in soils and become toxic to plants or micro-organisms and also present risks to aquatic ecosystems (see 3.3.b). Excessive concentrations in the soil can lead to yield reductions on some crops such as legumes and straw cereals (McGrath, 1981), with the free forms of Zn and Cu being the most phytotoxic (Suttle, 2010; Kumar *et al.*, 2021). Generally, pig manure is applied to soils assuming a N application rate of 170 kg.ha<sup>-1</sup>.an<sup>-1</sup>. This corresponds to Zn flows of about 2.38 kg.ha<sup>-1</sup>.year<sup>-1</sup> and Cu flows of about 1.53 kg.ha<sup>-1</sup>.year<sup>-1</sup>. These inputs far exceed the needs of the crops (Marcato, 2007) since it

is estimated, for example for wheat, that the plants export about 50 and 200 g.ha<sup>-1</sup>.an<sup>-1</sup> respectively. Moreover, the soil microbiota is also very sensitive to an increase in the content of TM. Zn and Cu once applied can exhibit different behaviours: they can accumulate in soils, be taken up by plants or be leached. This will depend on their speciation, the type of soil and the type of plant (Jondreville *et al.*, 2002; Legros, 2008).

#### a. Harvesting by plants

Zn and Cu are essential minerals for plant growth and are therefore taken up by plants. Plants contain between 10 and 100 mg.kg<sup>-1</sup> MB of Zn (Gräber *et al.*, 2005). They require on average between 5 and 20 mg.kg<sup>-1</sup> MB of Cu (Legros, 2008; Kumar *et al.*, 2021). Cu deficiency in plants leads to reduced plant growth, promotes leaf chlorosis and causes cytotoxicity (Kumar *et al.*, 2021). In some regions there is a risk of Zn and Cu deficiency (GIS Sol, 2001), with significant variability depending on the presence of pig or poultry farms and vineyards. Amendments rich in Zn and Cu may therefore be of interest. Pig manure has a higher Zn content than Cu, which leads to a more rapid increase in Zn than Cu content in soils (De Conti *et al.*, 2016). There is no risk of Cu contamination in the human food chain, as it is not very mobile in the plant and therefore rarely reaches the consumable parts of plants (Marcato, 2007).

#### b. Flows to groundwater

Zn and Cu accumulated in the soil after spreading can subsequently be leached and transferred to the water system. This depends on the speciation of these elements which will be more or less soluble depending on the physico-chemical characteristics of the environment. These transfers to aquatic ecosystems are also helped by erosion,

with Zn and Cu being transferred with soil or OM particles. In this way, Zn and Cu can be taken up by marine organisms and contribute to the contamination of the food chain (Jondreville *et al.*, 2002; Legros *et al.*, 2013).

#### c. Accumulation in soils

Many studies highlight that Cu is a much less accessible and less mobile element than Zn (Gräber *et al.*, 2005; Mallmann *et al.*, 2014; Marszałek *et al.*, 2019). Indeed, Cu has a higher affinity for OM (Kumar *et al.*, 2021) and is therefore more sensitive to soil properties such as OM content or pH. It can therefore become more or less mobile when these characteristics vary, but will mainly accumulate in the soil. Zn is also a few mobile element in soils, but it is less sensitive to variations in soil characteristics (Marszałek *et al.*, 2019). In the soil solution (water circulating in the open spaces of the soil), Zn will be mostly in the free form Zn<sup>2+</sup> while Cu will be mostly in the form complexed with OM (De Conti *et al.*, 2016; Kumar *et al.*, 2021).

Zn and Cu from applied livestock manure accumulate mostly in the topsoil (0-20 cm), i.e. the surface layer of soils (Jondreville *et al.*, 2002; Revy, 2003; Marszałek *et al.*, 2019). According to McGrath (1981), only 2-10% of the Cu brought in by land application is subsequently leached or taken up by plants. In soils that have received pig manure for many years, De Conti *et al.* (2016) observed an accumulation of carbon in the surface layer, having a high affinity with metals, in particular with Cu. This may explain the accumulation of these elements in this soil layer. Moreover, Zn and Cu are not very mobile and are therefore transferred in small quantities to the deeper soil layers. Zn and Cu can be transported by humic acid after formation of chelate complexes



and these complexes are more stable when formed with Cu than with Zn (Gräber *et al.*, 2005). This complexation of Zn and Cu in soils reduces the content of these elements in free form and thus their phytotoxicity (De Conti *et al.*, 2016). The soil pH is also a factor that has a strong influence on the solubility of Zn and Cu. Its increase improves the adsorption of minerals on OM and thus decreases their mobility (Levasseur, 1998; De Conti *et al.*, 2016). Acidic soils, as is often the case in Brittany, therefore have a higher solubility of Zn and Cu. Mallmann *et al.* (2014) assessed the effect of tillage practices on the distribution of Zn and Cu following pig manure inputs over a long period. They showed that no-till practices amplify the accumulation of Zn and Cu at the soil surface and reduce their mobility to depths exceeding 20 cm.

Many studies have evaluated the effect of long-term application of pig manure on Zn and Cu accumulation (Coppenet *et al.*, 1993; L'Herroux *et al.*, 1997; McGrath *et al.*, 2000; Gräber *et al.*, 2005; De Conti *et al.*, 2016; Formentini *et al.*, 2016; Benedet *et al.*, 2019; Benedet *et al.*, 2020). According to Coppenet *et al.* (1993), between 1973 and 1988, i.e. after 15 years of regular application of pig manure, the soils of Finistère (France) were enriched in Zn by nearly  $0.37 \text{ mg.kg}^{-1}.\text{an}^{-1}$  and in Cu by nearly  $0.22 \text{ mg.kg}^{-1}.\text{an}^{-1}$ . They also showed the more manure is applied the higher this increase is. L'Herroux *et al.* (1997) were interested in the speciation of Zn and Cu in the soils of Brittany (France), after 6 years of application of pig manure, and in their distribution in the different fractions determined according to their bioavailability: in the long term, Cu is mainly found in the poorly bioavailable fraction and with time the share of Zn increases in the slightly more bioavailable fractions. These observations on

the speciation of Zn and Cu or on their behaviour in soils are important to predict their effect on the environment (Formentini *et al.*, 2016). Indeed, the toxic effect of Cu in soils is a long-term environmental problem (López Alonso *et al.*, 2000). De Conti *et al.* (2016) evaluated this effect of Zn and Cu accumulation on soils and highlighted that the phytotoxic effect of Zn and Cu does not appear even after several years of pig manure application, because plants modify soil conditions so that the chemical species can complex with dissolved organic carbon. A certain stability of Zn and Cu is then observed. Plants generally tolerate Zn levels ranging from 10 to  $100 \text{ mg.kg}^{-1} \text{ DM}$  and Cu levels in soils ranging from 2 to  $40 \text{ mg.kg}^{-1} \text{ DM}$  (Gräber *et al.*, 2005). According to Coppenet *et al.* (1993), some Zn-sensitive plants, such as maize, can suffer from deficiencies in this element when the soil content is below  $1.5 \text{ mg.kg}^{-1} \text{ DM}$  and a phytotoxic effect is observed when this content exceeds  $120 \text{ mg.kg}^{-1} \text{ DM}$ . In Brittany soils, with a pH close to 6, this phytotoxic effect is observed when the accumulated content of Zn and Cu exceeds  $120 \text{ mg.kg}^{-1} \text{ DM}$ . Cu tends to accumulate in root tissues and can be transferred to the stem; Cu toxicity in plants thus affects root growth and plant morphology (Kumar *et al.*, 2021). McGrath *et al.* (1995) reported that soil microbial activity is disrupted when soil Zn levels exceed 100 to  $200 \text{ mg.kg}^{-1} \text{ DM}$ . Zhang *et al.* (2016) studied the effect of pig manure application on soil microbial community and put forward that Zn and Cu can improve soil fertility by allowing the development of soil biomass. According to them, Zn has the greatest influence on the soil microbiota.

The activity of earthworms in soils is reduced when the Cu concentration exceeds  $50 \text{ mg.kg}^{-1} \text{ DM}$  (Gräber *et al.*,

2005). Toxicity of Zn and Cu can also affect animal species after spreading pig manure on pastures. López Alonso *et al.* (2000) showed that in regions with intensive pig farming, more than 20% of the livestock have liver Cu concentrations exceeding the potentially toxic concentration of  $150 \text{ mg.kg}^{-1}$ . Sheep in particular are extremely sensitive to excess Cu and cases of poisoning of sheep grazing on grassland fertilised with Cu-rich pig manure have been reported (Poulsen, 1998).

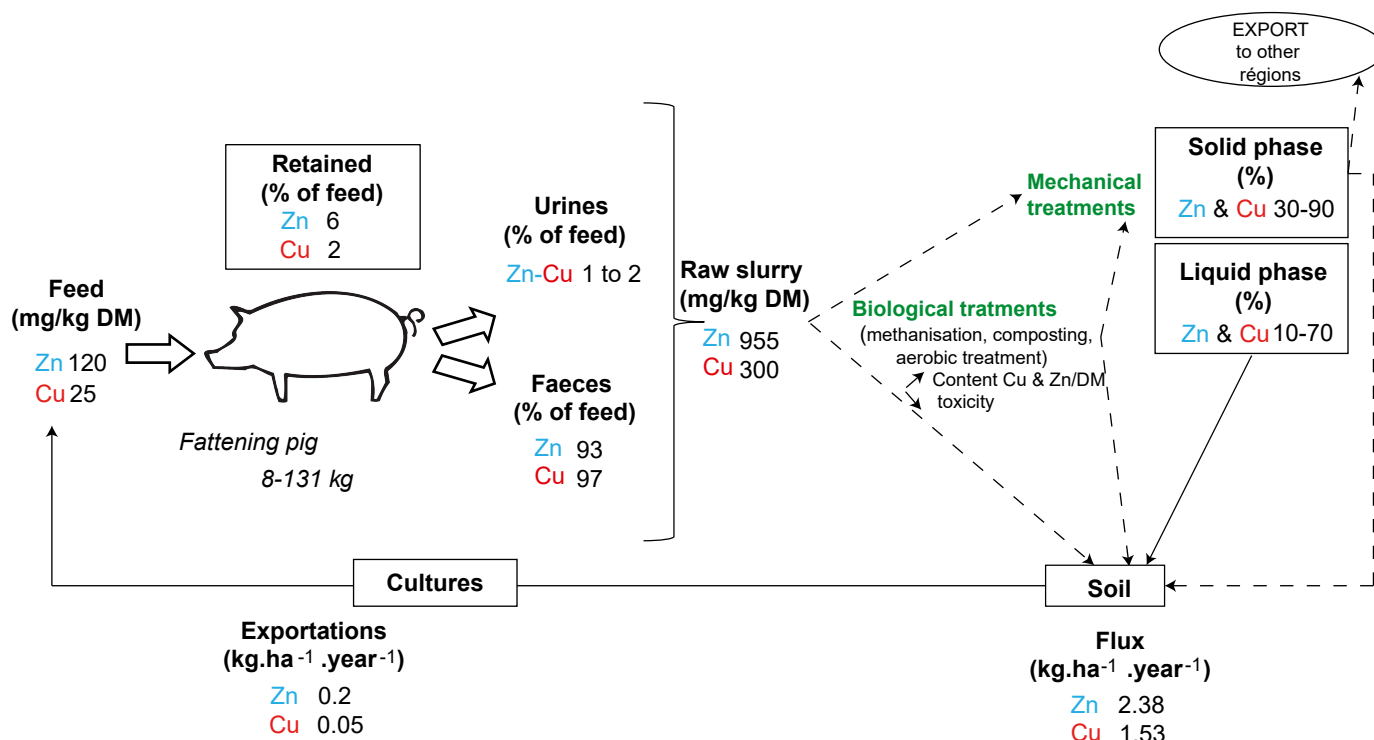
### ■ 3.4. Specific case of the return to the soil of products from the treatment of pig manure

As mentioned earlier, the treatment of pig manure does not remove Zn and Cu but concentrates them in some fractions. Møller *et al.* (2007) indicate that application of the solid phase from centrifugal phase separation results in a lower soil load of Zn and Cu than pure slurry, but this is probably very dependent on the generally N-based fertilisation rules. Studies evaluate the germination index of seeds after fertilisation with compost, which allows to determine the phytotoxicity and maturity of the latter. According to He *et al.* (2009), this index increases during the composting process: this means that this type of treatment leads to a decrease in phytotoxicity and improves root growth. Furthermore, this germination index can be predicted by the total content of Zn and Cu and other metals (lead).

## Conclusion

Zn and Cu are essential TM for pigs but potentially toxic for the environment, and they are limited resources. It is therefore essential to characterise their flow throughout the pig production

**Figure 7.** Copper (Cu) and zinc (Zn) flows through the pig industry and its effluent management systems. Example of fattening pigs. Circular bioeconomy approach.



chain in order to limit their use and application to agricultural land. Figure 7 summarises this feed-animal-manure-treatment-soil continuum in the context of the circular bioeconomy.

Reducing dietary Zn and Cu, while respecting the animal's requirement, is the main way to reduce the content of these two elements in pig excrement. This can be achieved by adapting inputs as closely as possible to changing needs and by using more bioavailable sources. Effluent treatment then allows better management of the return of Zn and Cu to the soil by modifying their speciation and their content in relation to the DM. Separative treatments allow

a redistribution of these elements between two phases and facilitate their export to regions with soils that are less rich in Cu and Zn, or even deficient, and on crops that have a high need.

### Authors' contributions

Emma Gourlez, Fabrice Beline, Jean-Yves Dourmad, Alessandra Monteiro, Francine de Quelen participated in the definition and content of the article. Emma Gourlez wrote the manuscript and considered the corrections made by the other authors. Francine de Quelen is the coordinator of the project funding this study, which was carried out as part

of a thesis directed by Fabrice Beline. All co-authors have read and approved the final version of the article.

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## Abstract

Zinc and copper are heavy metals that may have short- or long-term environmental risk for agricultural soils on which pig manure is spread. Zinc and copper are essential for pigs and farm performance, and they are added as supplements to the feed. Because of their low retention rate by animals, however, most of the amounts ingested are excreted in manure. Moreover, they are limited natural mineral resources. It is thus essential to characterize their flows better through the continuum of feed - manure - treatment - soils to manage their use better. A literature review of these flows indicates that adapting pig feed is the main mechanism available to decrease the release of zinc and copper into the environment. European Union regulations set maximum dietary concentrations of zinc and copper (e.g. 120 and 25 mg.kg<sup>-1</sup> DM for fattening pigs, respectively). Treating farm waste is another way to derive added value from these wastes and better control the transfer of zinc and copper to soils, by concentrating these elements in products that can be exported from pig farms to regions that need them. Thus, improved knowledge of their distribution through the production chain is necessary to manage their fate and evaluate their environmental risks better. More research is thus needed to refine feeding strategies that include zinc and copper, while maintaining animal performance and health and considering strategies of manure management and treatment, as part of the circular bioeconomy.

## Résumé

### Rôle et impact environnemental du cuivre et du zinc en élevage porcin : de l'alimentation au retour au sol des effluents

Le zinc et le cuivre sont des éléments-traces métalliques (ETM) qui peuvent présenter à plus ou moins long terme un risque environnemental pour les sols agricoles sur lesquels sont valorisées les déjections porcines. Ces ETM sont essentiels pour la croissance et la santé des porcs en élevage, ils sont donc ajoutés dans l'aliment sous forme de supplémentation. Cependant, du fait de leur faible taux de rétention chez le porc, ils se retrouvent très majoritairement excrétés dans les déjections. De plus, ce sont des ressources minérales naturelles limitées. Il est alors essentiel de caractériser leurs flux le long du continuum aliments-déjections-traitements-sols afin de maîtriser au mieux leur utilisation. Un état des lieux des connaissances sur ces flux montre que l'alimentation est le principal levier pour limiter les rejets de ces ETM vers l'environnement. La réglementation européenne fixe actuellement des teneurs limites des aliments en zinc et en cuivre (e.g. pour un porc en engraissement, 120 et 25 mg.kg<sup>-1</sup> MS respectivement). Le traitement des déjections est un second levier pour optimiser la valorisation de ces effluents d'élevage et mieux gérer la redistribution de ces ETM sur les sols, en les concentrant dans certains produits plus facilement exportables vers des zones en déficit. Une meilleure connaissance de leur forme chimique tout au long de la filière est aussi nécessaire pour mieux maîtriser leur devenir et préciser les risques pour l'environnement. Des recherches restent nécessaires pour affiner les stratégies d'apport dans les aliments, tout en conservant les performances et la santé des animaux et en prenant en compte les différentes modalités de gestion des effluents dans le cadre d'une bioéconomie circulaire.

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