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REVIEW

The current phosphate recycling situation in China and Germany: a comparative review

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Abstract Phosphorus (P) is an indispensable element for organisms but the primary source of P—mineral phosphate resources—are non-renewable. Agricultural production has a high demand for fossil phosphate resources, but the resulting phosphate-rich residues are lack of management. This leads to rapid reserves depletion and severe phosphate pollution risks. One sustainable way is to reuse the phosphate dispersed in various residues such as sewage sludge and livestock manure. Diverse techniques have emerged to recover phosphate from wastes to close the phosphate cycle. While it is a global issue, the regional situations regarding potential phosphate scarcity and its management differ strongly. China is rich in phosphate resources, but over-exploitation has greatly increased the risk of phosphate rocks depletion, while in Germany the P resources depend on imports, but there is commitment to keep a balance between import and utilization. This had led to great differences in the way the two countries deal with the "re-use" of phosphate in waste. China is now in a transition phase from the simple terminal pollution control to "waste" reuse and nutrient resources recycling. One sign of this tendency is the mandatory garbage classification and preparation for further processing and recycling. This was first implemented in Shanghai in 2019, whereas Germany has been following the legal framework for waste management since the 19th century. There are a series of laws to control the nutrient loss from municipal and agricultural activities, as for instance with sewage sludge ordinance and fertilizer legislation. Many of these laws have been newly revised recently. Sewage sludge cannot be directly utilized on farmland as organic fertilizer any more. Alternatively, phosphate and other nutrients should be recovered from sewage sludge. Advanced phosphate recovery technologies and related nutrient recycling schemes are proceeding. This review summarizes the current situation of phosphate-containing residues management and phosphate reuse in China and Germany. The state legislation and policies, which would affect the phosphate recycling concept are presented as well. As there are various kinds of phosphate-containing residues, different phosphate recovery technologies can be applied. Those technologies are discussed from their mechanism and suitability.

Keywords phosphate recovery, manure, sewage sludge, ordinances, technologies

1 Introduction

Phosphorus (P) is an indispensable and irreplaceable element for all living matter on earth[1,2]. As phosphate, it is an important component of ATP (adenosine triphosphate)—the energy source for cellular activities[3,4]. Phosphate-based products are widely used in agricultural production, e.g., fertilizer, pesticide, and animal feed[5,6]. The corresponding production relies on the raw material phosphate, which is mainly of fossil origin[7].

The primary source of P is phosphate rock mining, and the deposits are mainly located in several countries[8]. Nowadays, agricultural production is still highly dependent on mineral phosphate. Among the global phosphate rock production over 86 wt.% are used for mineral fertilizer, 10 wt.% contribute to food additives, while a small portion (about 4 wt.%) is used for the synthesis of chemicals[9]. The prediction about how many years the current phosphate reserves can afford those productions varies between 100 and 400 years[10]. However, it is worth noting that along with the growing world population, the

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*These authors contribute equally to the work increasing demand for animal and biofuel production, the phosphate requirement is also increasing by 2.5%–3% per year[11,12]. It is likely the remaining phosphate resources will be depleted in the foreseeable future.

While the global phosphate resources crisis is getting worse, the phosphate utilization efficiency (PUE) in agricultural production remains quite low. The PUE can be expressed as grain yield over available phosphate fertilizer per unit soil. Only around 16 wt.% of phosphate in fertilizers flows to food along with the crop harvest, while the rest remains in the soil, crop residues and ends up in animal manure[13]. A small portion of the phosphate in food can be assimilated by humans while the remainder is collected in wastewater treatment plants (WWTPs)[14]. The ecological system is suffering from the excessive phosphate, namely, surplus of phosphate in farmland and eutrophication of water bodies.

In this situation, the contradiction between mineral phosphate resources utilization and P-rich residues management intensifies: on one hand is the increasing demand for phosphate resource with the rise of grain and meat production; on the other hand, much more phosphate is lost with agricultural residues, further deteriorating the eutrophication situation. A sustainable way to reconcile this contradiction is to come up with a feasible phosphate-recycling scheme. This is not only crucial for those countries with scanty phosphate rock reserves that relying on its import, such as Germany, but also for countries who have over exploited phosphate reserves or are faced with severe phosphate pollutions, such as China.

Phosphate can be recovered from various kinds of feedstocks, such as sewage sludge from WWTPs, livestock manure, agri-food residues, etc.[15–18]. Regarding the various feedstocks and their different phosphate morphologies and distributions, an abundance of phosphate recovery methods have been developed. Struvite crystallization is an effective approach to recover magnesium ammonium phosphate from streams containing aqueous phosphate[19]. Wet chemical leaching is a common means to elute phosphate from solid phase to acidic or alkaline solutions[20]. Thermochemical process can be conducted in thermal or hydrothermal ways, by which phosphate bound by heavy metal ions are released at elevated temperatures[21].

The motivation to develop phosphate recovery processes heavily depends on local mineral phosphate-rock resources, animal husbandry methods, state regulations concerning fertilization and waste management. In Germany and most other European countries, which have no phosphate reserves, most of the phosphate resources are imported. These countries have long experience of phosphate recovery as they have always been trying to reduce their dependence on imports. China is rich in phosphate deposits. As greater strategic value has begun to be placed on the phosphate resources, greater importance has been attached to them and China is most likely to tighten domestic phosphate mining, curtail exports[22] and to prompt phosphate recovery.

Moreover, Germany is faced with stricter standards concerning the agricultural use of sewage sludge and China has been aware of negative effects of improper phosphate resources utilization. It is therefore mutually beneficial to carry out a joint research regarding the reuse of P. Both sides are putting effort into developing advanced phosphate recycling concepts for practical application.

This review aims to arouse the public awareness of the situation of phosphate rock resources, which is not so optimistic: there is a dramatic phosphate sink with agricultural residues and municipal waste disposal. Under such circumstances, phosphate recycling is necessary and pressing. The relative legal framework has its enforcement function to push the phosphate recycling forward. The sewage sludge ordinance and fertiliser legislation which exist in Germany and China are discussed. Germany has fertiliser regulations that strictly limit the nitrogen (N) and phosphate emission from farming systems to water bodies, as well as a sewage sludge ordinance that explicitly stipulates the nutrient recovery from sewage sludge. By contrast, phosphate recovery in China is still lacking in support from relative legislations. This review will draw the attention of Chinese decision makers to the phosphate loss. Moreover, phosphate recovery technologies play a key role in achieving phosphate recycling. Therefore, this review presents the state of the art technologies for phosphate recovery. Due to the operating cost or technical bottlenecks of scale-up, some of the phosphate conceptual designs are only available at a laboratory- or pilot-scale. Joint research between China and Germany could identify more possibilities for technologies development and application. Common progress in the two countries is expected.

2 Current situations of P utilization

2.1 Phosphate rock reserves and mining

According to the latest report released by USGS (United States Geological Survey)[8]: the global phosphate rock resources, including the storage on the continental shelves and in the oceans, is around 300 billion tons. Only 70 billion proved to be recoverable and these resources are mainly situated in Morocco and Western Sahara (71%), China (5%) and Algeria (3%) (Table 1).

As the largest producing area of phosphate deposits, Morocco and Western Sahara have a relatively low mining capacity. By contrast, China, which only possesses 5% of the global phosphate rock resources, has achieved more than 53% of the worldwide production capacity, which is almost five-times the production rate of the ‘phosphate-rich’ countries. This example reveals that global phosphate resources are not distributed in the areas of intensive demand. There are still many problems in the development and utilization of phosphate rock in China. 70%–80% of the total phosphate rocks are sedimentary and thus need to be exploited underground with lots of difficulties[23]. In addition, according to the grade classification, the average grade of phosphate ore is only 18 wt.% of P_2O_5. Except for the exported fraction, most of the high and middle grade (high grade: P_2O_5 content > 30 wt.%, middle grade: 30 wt.% > P_2O_5 > 26 wt.%) phosphate rock (> 80.5 wt.% of total production) is mined and processed into phosphate fertilizer for agricultural production in China, to supply food for 22% of the global population[24,25]. However, if China maintains this annual rate of production, the high-middle grade phosphate rock resources will run out in less than a decade[26,27].

Unlike China, European countries have no abundant phosphate rock resources. In the European Union (EU) only Finland has phosphate rock resources (1 billion tons) and active mining (0.95 million tons per year) (Table 1), and only 10 wt.% of the EU P demand can be supplied by Finland. Although the phosphate content in soil is abundant, especially in Germany, in order to ensure agricultural production, Europe still needs to rely on phosphate imports[28]. Hence, the European Commission has listed P as a critical raw element since 2014[29].

Table 1 Phosphate rock reserves and production, 2017[8]

Site	Reserves (billion tons)	Production (million tons)
Morocco and Sahara	50	27
China	3.3	140
Algeria	2.2	1.3
United States	1.0	27.7
Russia	0.7	12.5
Finland	1.0	0.95
Worldwide	70	263

Furthermore, the mining of phosphate rock has many problems, for example: (1) a fair portion of the existing phosphate rock resources is below the acceptable quality needed for fertilization; (2) CO_2 and particle matter emissions during the mining process would intensify climate change and air contamination[30]; (3) unbalanced distribution of phosphate rock mass, i.e., regional separation of P production and consumption[5]; (4) the mining leads to a regional high contamination with heavy metals[31].

Due to the exploitability of phosphate rock, China has chosen to reduce the exports amount to secure its domestic supply[28] instead of increasing imports. Closing the P circle is a key aspect for the sustainable usage of the remaining resources and also a way to be less dependent on imports. Therefore, Germany as well as the EU, set certain guidelines concerning phosphate recovery, which have to be met in the future.

2.2 Renewable phosphate resources

A considerable amount of phosphate is consumed as food or feed by humans and animals, respectively. Most of this phosphate cannot be absorbed and is excreted by living organisms[11]. This is the reason why a lot of phosphate accumulates in animal manure, municipal sewage[2,32] as shown in Table 2. P does not disappear but instead flows into the environment, which will gradually sharpen the contradictions between dwindling phosphate resources and damage to the ecological environment[33]. The utilization of these waste streams as renewable P resources is considered as well as to decrease phosphate rock mining and therefore alleviate the consequential environmental problems.

Germany and China have both applied nutrient recycling from agri-/non agri-residues back to crop cultivation in their development process for farming and animal husbandry[34]. The general phosphate recycling concept is shown in Fig. 1. Sewage sludge and livestock manure are selected as the two typical renewable phosphate resources. Sewage sludge is derived from urban areas, containing human excreta, domestic wastewater, etc., which is treated in WWTPs. The phosphate recycling can be achieved by reuse of certified sewage sludge on farmland, phosphate recovery from incinerated sludge ash, and struvite precipitation from the aqueous phase.

Livestock manure is collected in rural intensive livestock and poultry farms. In many farms, the solid-liquid separation is applied on manure first, via which most of the phosphate is concentrated in solid phase. The solid phase can be directly used as fertilizers. Alternatively, it can be further processed by means of composting or thermal treatment. There is also the option of liquid-solid separation of digestate after biogas fermentation, i.e., anaerobic digestion, and then composting the solids.

As there are many alternatives for phosphate recycling, there are differences in Germany and China in phosphate fertilizers production (or import) and consumption, national policies and corresponding technologies development. These factors in the two countries are worthy of discussion.

Table 2 Phosphorous contents in different wastes (on dry matter), Europe and China

Site	Animal manure	Sewage sludge	Animal by-product	Food residue
Europe ($kt \cdot yr^{-1}$ P)[a]	1810	374	312	187
China ($kt \cdot yr^{-1}$ P)[b]	> 2000	> 200	> 200	> 200

Note: [a] Data from Nättorp et al.[34]; [b]data from Zheng et al.[32].

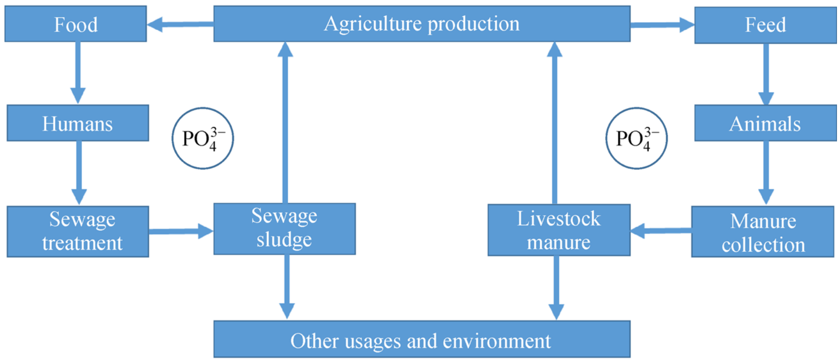

Fig. 1 Phosphate flow between agriculture production and environment (adapted from Nättorp et al.[34]).

2.3 Current situation of phosphate reuse

2.3.1 Livestock manure generation and utilization

In pre-industrial times, phosphate used for agricultural production was mainly recycled from human excrement and livestock and poultry manure as organic fertilizer[35]. Due to population increase and farming system diversification, the mineral fertilizers began to dominate the agricultural production to enhance crop yields. In the meantime, economic development and urbanization have had a significant impact on the pattern of food consumption, especially regarding the demand for protein from animals[36]. This has led to a sharp increase of livestock production and resulted in a large amount of produced manure[37]. Whiles this trend started earlier (first urban transition, 1750–1950) in Europe than China (second urban transition, 1950–2030)[38], the current development in policies also differ as shown in the following.

2.3.1.1 China

In 2010, the First National Pollution Source Census Bulletin (China) found that livestock farms had become one of the great contributors to anthropogenic P losses[39]. According to the official data from the Chinese Ministry of Agriculture and Rural Affairs (MARA) in 2016, the annual production of livestock and poultry manure was about 3.8 billion tons[40]. After estimates, these manures contain 14.2 million tons of N and 2.5 million tons of P[41]. It is worth noting that these wastes, which could be important materials for anaerobic digestion (AD) biogas plants and application on farmland as organic fertilizers, may also be a serious source of pollution if not properly treated before being discharged into the environment. This has received increasing attention from China's decision-makers/stakeholders[42].

From 2003 to 2015, 110975 biogas projects were completed in China with the support of central and local investment, 99.6% of which were livestock and poultry manure as the main raw material[43]. However, it should be realized that, despite the large number of biogas plants constructed in China since 2003, many of these plants are not managed well and therefore not working properly, especially small and medium-sale ones. Although a "Renewable Energy Law" has been issued in China since 2006, it is hard for the biogas operations to sell the electricity generated to the State Grid. The potential amount of livestock and poultry manure consumed for biogas projects exceeds 1 billion tons[41], and the rest of the manure should be composted or directly used as fertilizer. But according to the P flow analysis, less than 50 wt.% of the manure or the manure products were recycled to arable land[15,44]. In fact, the comprehensive utilization ratio of manure was less than 60%[45].

China's decision-makers have begun to encourage the reuse of manure wastes through technological progress, because it constitutes a potential way to achieve economic and environmental benefits[46].

2.3.1.2 Germany

Livestock production is also an important part of German agriculture. The output value of animal husbandry accounts for 62.14% of the total German agricultural output value[47]. Germany is one of the top ten global producers concerning pig and cattle meat (Table 3). It was estimated that 139 million tons of manure are produced each year[48] and this large amount increases the risk of uncontrolled nutrient loss into the environment[49]. There are a few possible ways to resolve this problem and produce valuable products at the same time; one of them is the production biogas from manure[50]. Projects concerning biogas started in the early 1990s, back then there were only 139 in the country. Since the implementation of the "Renewable Energy Law" in 2000, biogas projects have been strongly promoted through demonstration projects, resulting in an increase of 1450 projects by 2004. Until 2018, 9500 biogas projects had been completed[51]. Unlike the situation in China, most biogas plants constructed in

Table 3 Production of pig and cattle meat indigenous[47]

Top 10 producers	Production of pig meat indigenous (million tons)	Top 10 producers	Production of cattle meat indigenous (million tons)
China	821	United States	237
United States	186	Brazil	158.3
Germany	84.8	China	99.7
Spain	62.5	Argentina	58.7
Brazil	56.4	Australia	46.5
France	46.9	Russian	43.3
Canada	45.6	France	38.7
Viet Nam	41.7	Mexico	36.6
Netherlands	40.1	Canada	29.9
Poland	39.8	Germany	29.3

Germany since 2000 are mainly operating with biogas silage maize while only a small portion is conducting co-fermentation of silage maize with livestock manure. The number of newly-built biogas plants has sharply declined since the revision of the Renewable Energy Law in Germany in 2017.

In addition, farms adopted a kind of manure disposal system, which centralizes the feces and urine in the cesspool under the barn and with a storage capacity of 6–9 months. After maturity, it is directly returned to the field. Germany is a country with a lack of energy and phosphate resources. Therefore, the government committed itself to supporting the development of renewable technologies. The biogas engineering is more encouraged to solve the problem of manure disposal. Nowadays, Germany has reduced P losses in the agricultural sector due to a 97 wt.% manure P-recycling ratio[52].

2.3.2 Sewage sludge generation and utilization

With the increase in urbanization, the construction of sewage treatment facilities has developed rapidly. The construction and operation of sewage treatment plants has achieved the reduction of sewage and the control of pollution but produced a large amount of sludge[53]. After the application of chemical and/or biological phosphate removal in WWTPs, 90 wt.% of the total P is contained in the sludge[54], which therefore could be considered as the most abundant renewable P resources.

2.3.2.1 China

According to the official data, 18 million tons of municipal sludge (80 wt.% water content) were generated in 2018, of which, 26.5 wt.% were used directly as organic fertilizer, 14.4 wt.% for building materials, 25.0 wt.% for incineration, 24.1wt.% for sanitary landfill, and about 9.3 wt.% were treated by other comprehensive methods (Fig. 2)[55]. However, two problems should be kept in mind. First, there is the policy about the reuse of sludge, which must be treated in a non-hazardous way before it is applied on the field. The non-hazardous treatment aims to eliminate microorganisms and pathogens. A long-term implementation of the land filling method is hindered by the bearing capacity of the soil. Second, although the landfill method may be effective, it is not a solution for the P depletion crisis.

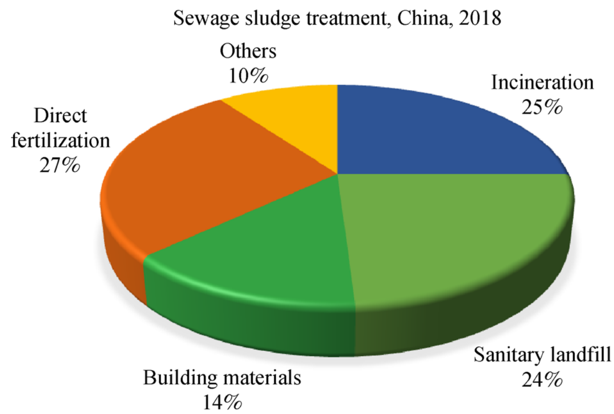

Fig. 2 Sewage sludge treatment in China, 2018.

2.3.2.2 Germany

According to a recent survey of Germany, around 1.8 million tons of dry sludge were produced from sewage treatment plants in 2016[56]. Figure 3 shows the distribution of the designated uses for sewage sludge in Germany. Around 24 wt.% are used for fertilization purposes while nearly 65 wt.% of the sewage sludge is incinerated. However, the former usage causes soil contamination due to the heavy metals and organic pollutants contained in the sewage sludge[57], while the latter leads to a significant loss of reusable phosphate. The percentages of these two methods will further decline in the near future. Previous case studies concerning phosphate recovery from sewage

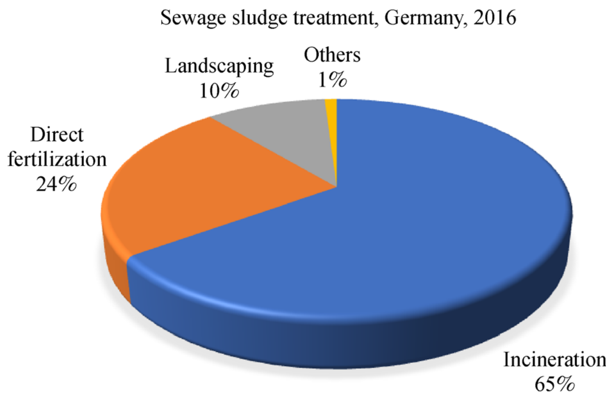

Fig. 3 Sewage sludge treatment in Germany, 2016.

sludge were of the ancillary works for the removal of heavy metals, because Germany has the strictest fertilizer regulations. The German Federal Environment Agency (UBA) has already started to enforce phosphate recovery in WWTPs.

The proportion of sludge incineration in Germany is much higher than in China. A large proportion of sewage sludge in China has been used for sanitary landfill. The contamination risk varies depending on the different treatment methods, but China may already face a serious problem. Since sewage sludge is rich in phosphate, Germany shifted the focus of its sewage sludge management strategies from pollution reduction to resource recovery. China on the other hand is currently still at the stage of pollution control.

3 State ordinances for P recovery

For quite a long time, phosphate reuse was achieved by applying organic phosphate-rich residues on farmland. However, the organic phosphate has a relative low solubility and bioavailability in soil. The better route is to recover inorganic phosphate by means of a biomass refinery processes. The resulting inorganic phosphate fertilizers are more efficient for agricultural production.

The new German sewage sludge ordinance entered into force in October 2017 clearly emphasized the phosphate recovery from sewage sludge. This is a global sign that the inorganic phosphate extraction will replace the conventional phosphate reuse.

3.1 The ordinances of P recovery in Germany

3.1.1 Manure

Residues from intensive livestock production systems, e.g., livestock manure and biogas digestates, contain a considerable amount of nutrient elements such as P and N compounds. In Germany, the Fertilizer Ordinance (FO)[58] restricts the organic fertilizers utilization and nutrient loss from these organic sources to the environment. The German FO has been newly revised in 2017 and specifies the fertilizer planning, N application threshold of manure, nutrient balances, manure storage capacity and application, etc.

The fertilizer planning, chemical and organic fertilizer utilization must relate to crop yields, nutrient content of manure and nutrients in soil. The application of organic nutrients on farmland is quantified by mineral fertilizer equivalents (MFE). MFE are the nutrient contents in the organic fertilizers that can replace the application of mineral fertilizers. For instance, the N source from swine and cattle manure can afford 50% and 60% of the plant demand, respectively[58]. In German Fertilizer Action, nutrient balances indicate the threat of nutrients loss to water bodies. An overview of the nutrient balance is illustrated in Fig. 4.

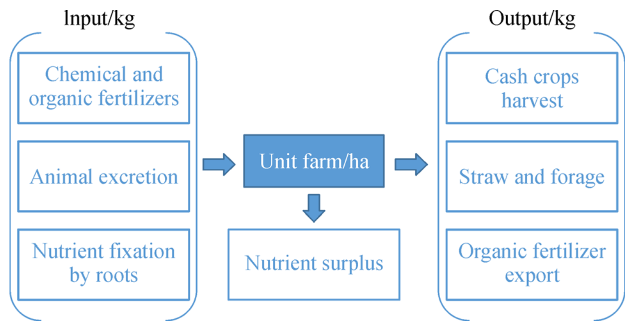

Fig. 4 Nutrient balances in unit farmland (adapted from Kuhn[58]).

The EU stipulates an upper limit for N content of organic fertilizers spread to arable farmland, which is 170 $kg \cdot ha^{-1} \cdot yr^{-1}$ N. The German FO previously only considered animal-derived organic fertilizers to be managed by this threshold. The revised German FO (2017) now also includes biogas digestates derived from biogas silage maize under the 170 $kg \cdot ha^{-1} \cdot yr^{-1}$ N. Although there is no express provision for P planning in the FO 17, phosphate fertilizers application should be carefully calculated based on the nutrient balance. It is necessary to conduct soil analysis before fertilization. Phosphate fertilizers application on soil with more than 20 mg P_2O_5 per 100 g is limited to demand of less than 60 $kg \cdot ha^{-1}$ P_2O_5[57].

The nutrient surplus is restricted as the allowed surplus of N is 50 $kg \cdot ha^{-1} \cdot yr^{-1}$ (average value over a three-year period), while the allowed surplus of P is 10 $kg \cdot ha^{-1} \cdot yr^{-1}$ (average value over a six-year period)[58].

In the FO 17, the application of organic fertilizers application from December 15 to January 1 is forbidden. This is defined as a blocking period. Additionally, manure application technologies are specified in FO 17 and the spreading way of organic fertilizer spreading is forbidden.

Instead, methods such as injection and drag shoe are permitted, in order to introduce organic fertilizers into the soil directly.

3.1.2 Sewage sludge

There are several laws and enactments regarding sewage sludge usage in Germany, which must be obeyed. These enactments can be differentiated in terms of their main objective. The sewage sludge ordinance and EU-sewage sludge directive focus on all applications of sewage sludge and its derivatives as well as the recycling of phosphate.

One objective of the sewage sludge ordinance is the regulation of the application of sewage sludge and all its derived mixtures and composts on soil, in agreement with the different existing laws concerning fertilizer. The distribution of phosphate is only allowed on certain agricultural fields, which fulfil a list of explicitly defined requirements[59]. Due to the tightening of the laws concerning sewage sludge the application on farmland will decrease and the thermal application will further increase. Due the latest amendment (2017) it will be mandatory, for the first time, for waste water treatment plants to install a phosphate recovery system for thermally used sewage sludge and the ashes thereby derived. The starting date for the mandatory phosphate recovery is linked to the size of the waste water treatment plant. Sewage sludge must be introduced to a recovery system if the P content is above 20 g·kg^{-1} DM). The executed recovery system should lower the P content either by 50% or below the maximum value of 20 g·kg^{-1} DM). If a thermal process is applied prior to the P recovery the chosen recovery system for the resulting ash or carbon material must be able to reduce the contained P by 80 wt.% or below the value of 20 g·kg^{-1} DM[58]. The storage of sewage sludge or its ashes is only allowed if mixing and losses are avoided and a P recovery in the future is ensured. Lastly, the incineration of sewage sludge will only be allowed with gas and coal.

Sewage sludge and all deriving products like ash or recycled products from phosphate recovery are defined as phosphate fertilizer by the German FO. The combined applicable threshold values for minor components of the sewage sludge ordinance and the fertilizer regulation for sewage sludge are given in Table 4[58–60].

3.2 The ordinances of phosphate recovery in China

3.2.1 Manure

From scattered raising to intensive livestock and poultry farming, the discharge of animal-source wastewaters or manure-derived products to cropland has always been an important manure disposal route in China[61,62]. The manure can be discharged as sewage (complying with GB5084-2005 and GB18596-2001), or it can be used after being rendered non-hazardous by treatment, as stated in GB18596-2001. Non-hazardous treatment refers to the use of high temperatures, aerobic or anaerobic technologies to kill pathogens, parasites and weed seeds in livestock and poultry manure (NY/T 1168-2006). The obtained organic fertilizer product or biogas slurry fertilizer must be applied according to the corresponding standards as illustrated in Table 5. In addition to specifying how to apply fertilizers to different types of soil (dose, season, etc.), these standards also require the control of fertilizer quality (Table 6). However, the current mismatch between the generated amount of manure and the treatment capacities has caused a serious ecological crisis.

In 2015, several documents were promulgated by the Central Government and the State Council, aiming to solve the outstanding problems such as the polluted environment in rural areas and the treatment of wastes like manure[63]. In 2017, a major strategy concerning the “rural revitalization” was put forward, including the implementation of rural green development methods. Additionally, guiding document on the disposal and utilization of livestock and poultry waste were also issued for the first time in China[64]. Furthermore, the action plan of livestock and poultry manure resource utilization (2017–2020) has set the following targets: “the utilization rate of livestock and poultry manure in China reaches above 75%, and the matching rate of treatment facilities and equipment are above 95%”. In 2018, the action plan for the fight against agricultural pollution in agriculture and rural areas began to be implemented[65].

The full-scale livestock and poultry breeding industries are the main targets for N as well as phosphate pollution prevention and control and have received the attention of the government and all relevant departments. In these guidance documents, composting as well as anaerobic digestion remain the main treatments, which farmers are

Table 4 The obligated threshold values, which have to be met concerning the application of sewage sludge and all its derivatives on soil in Germany[58–60]

Application	Cd	Hg	Pb	Cr	As	Cu	Zn	Ni	Fe	Organic compounds	Hygienic standard
Fertilizer regulation (mg·kg^{-1} DM)	1.5	1	150	2	40	900		80		√	√
Sewage sludge ordinance (mg·kg^{-1} DM)				Measure			4000		Measure	√	√

Table 5 Laws and ordinances for the treatment and application of manure in China

Law, standards and specifications	Code	Effective from
Irrigation water quality		
Standards for irrigation water quality	GB5084-2005	2006
Discharge		
Discharge standard of pollutants for livestock and poultry breeding	GB18596-2001	2003
Non-hazardous treatment		
Technical requirement for non-hazardous treatment of animal manure	NY/T 1168-2006	2006
Technical requirement for non-hazardous treatment of biogas slurry used in agriculture	Under consultation-2017	
Products		
Organic-inorganic compound fertilizers	GB18877-2009	2012
Organic fertilizer	NY525-2012	2012
Compound microbial fertilizers	NY/T 798-2004	2004
Microbial organic fertilizers	NY/T 884-2012	2012
Application		
Technology code for land application rates of livestock and poultry manure	GB/T 25246-2010	2011

Table 6 The obligated threshold values of the application of manure and all its derivatives on soil, China

Code	NH_4^+-N	P	Hg	Cd	As	Cr	Pb	Cu	Zn		Hygienic standard
GB5084-2005/($mg \cdot L^{-1}$)			0.001	0.01	0.05–0.1	0.1	0.2	0.5–1	2		√
GB18596-2001/($mg \cdot L^{-1} \cdot d^{-1}$)	80	8.0									√
NY/T1168-2006											√
GB18877-2009 ($mg \cdot kg^{-1}$ DM)	N, P, K $\geqslant$150000 (I)		5	10	50	500	150				√
NY525-2012 ($mg \cdot kg^{-1}$ DM)	N, P, K $\geqslant$50000		2	3	15	150	50				√
NY/T798-2004 ($mg \cdot kg^{-1}$ DM)	N, P, K $\geqslant$40000		5	10	75	150	100				√
NY/T884-2012 ($mg \cdot kg^{-1}$ DM)			2	3	15	150	50				√
GB/T25246-2010 ($mg \cdot kg^{-1}$ DM)					30–50 30–50 30–50			85–400 170–800 170–800	500–2000 700–2700 900–3400	<6.5 6.5–7.5 >7.5	√

Note: Hygienic standards include the mortality of aphid eggs, number of *E. coli*, etc.

recommended and encouraged to use. However, it is difficult to remove or heavy metals with those non-hazardous treatment techniques. Therefore, because of the large amount of continually generated manure it is impossible to only use non-hazardous treatment technologies for pollutant reduction. Consequently, the development of new technologies is crucial. In addition, while China's overall planning takes account of pollution control, there is no specific policy or plan for phosphate recycling. Problems like the low utilization efficiency of phosphate in animal husbandry and the pollution caused by manure will, it is hoped, be improved by the implementation of these policies.

The supernatant after manure treatment shall be in accordance with the regulation of GB5084-2005 as irrigation water for farmland. And if it is discharged in the form of sewage, it shall comply with the regulation of GB18596-2006.

3.2.2 Sewage sludge

Land use of sewage sludge mainly concerns land improvement and landscaping, etc., while sludge which meets certain standards (such as GB 4284-2018) is permitted for restricted agricultural use. The quality of sewage sludge produced from municipal wastewater treatment plant should meet the following requirements, shown in Table 7. Agricultural application of sewage sludge of course demands higher quality requirements than

Table 7 The obligated threshold values of the application of sewage sludge on soil, China

Application		Cd	Hg	Pb	Cr	As	Cu	Zn	Ni	Hygienic standard
Sludge treatment										
GB 24188-2009 ($mg \cdot kg^{-1}$ DM)	A	3	3	300	500	30	500	1200	100	√
	B	15	15	1000	1000	75	1500	3000	200	
Land use										
GB 4284-2018 ($mg \cdot kg^{-1}$ DM)	Acid soil	< 5	< 5	< 300	< 600	< 75	< 100			√
	Alkali soil	< 20	< 15	< 1000	< 1000	< 75	< 200			
GB/T 23486-2009 ($mg \cdot kg^{-1}$ DM)	Acid soil	< 5	< 5	< 300	< 600	< 75	< 800	< 2000	< 100	√
	Alkali soil	< 20	< 15	< 1000	< 1000	< 75	< 1500	< 4000	< 200	
GB/T 24600-2009 ($mg \cdot kg^{-1}$ DM)	Acid soil	< 5	< 5	< 300	< 600	< 75	< 800	< 2000	< 100	√
	Alkali soil	< 20	< 15	< 1000	< 1000	< 75	< 1500	< 4000	< 200	

Note: Only the A degree means it could be used to the plants for food product.

the other land use methods (Table 8). However, the problem of "attaching importance to sewage but ignoring sludge" is prominent in China. Insufficient attention and investment have been paid to the construction of sewage sludge treatment and disposal facilities, and consequently only a small proportion of sewage sludge is used for agriculture application. Furthermore, in the actual application process, most of the sewage sludge has not been treated or properly disposed, causing China to be confronted with severe environmental problems and health challenges[56]. According to the results determined by Ren et al.[66] the most important barriers that hinder the sustainable development of sludge-to-energy is the lack of project experience and technological immaturity. Recently, China has been attaching great importance to the construction of an "ecological civilization". The comprehensive work plan for energy conservation and emission reduction states that sewage sludge should be treated in a safe way and secondary pollution should be prevented[67]. Thus, it is necessary to enhance the project experience on sludge treatment by inviting foreign investors or technology providers to participate in the technology enhancement of sludge conversion into energy in China.

3.3 Ordinances-guided phosphate recovery trends in China and Germany

As discussed above, for manure phosphate treatment in China, there is a delay in the ordinance implementation, and there are many practical barriers and constraints. The "blocking periods (Germany)" concept is worthy of reference. For example, during the execution time, there may be an asynchronous phenomenon in municipalities, counties, and villages at all levels. Efforts may occur to improve the technologies in the current pilot area, and at the same time reserve "blocking periods" occur for non-pilots. Furthermore, it has been observed that a combination of measures in phosphate recovery is much more effective than a single one. Both in China and Germany at present, the farmland reuse is the main treatment measure for P utilization of livestock and poultry manure. Phosphate recovery is a global process, not only driven by local regulations, but also motivated by the development of relevant technologies. The introduction of industrial technologies into the agricultural sector is, however, a huge challenge.

For the sewage sludge phosphate treatment, the "mandatory phosphate recovery" strategy executed in

Table 8 Laws and ordinances for the treatment and application of sewage sludge in China

Law, standards and specifications	Code	Effective from
Sludge quality		
Quality of sludge from municipal wastewater treatment plant	GB 24188-2009	2010
Control standard		
Control standards for pollutants in sludges for agricultural use	GB 4284-2018	2019
Disposal ways		
Disposal of sludge from municipal wastewater treatment plant—quality of sludge used in gardens or parks	GB/T 23486-2009	2009
Disposal of sludge from municipal wastewater treatment plant—quality of sludge used in land improvement	GB/T 24600-2009	2009

Germany can be used as a typical demonstration. The central and local governments of China will need to take an active role in raising overall technical effectiveness about waste phosphate minimization. Improving sustainability performance should be an important selection criterion of technologies by the decisionmakers/stakeholders. It is worth mentioning that the recovered inorganic fertilizers have a higher solubility in soil and, therefore, greater plant availability than organic fertilizers. Furthermore, the transportation costs can be reduced, since inorganic fertilizers are less bulky than sludge or biogas digestates. Under such circumstances, countries which still lack ordinances concerning phosphate recovery, e.g., China, have already taken action and developed phosphate recovery technologies and been assessing their feasibilities in industrial applications.

4 Phosphate recovery strategies

By means of conventional nutrient recycling (i.e., organic recycling) (Fig. 1), derivatives resulting from sewage sludge and livestock manure treatment can also be utilized for further phosphate recovery. For instance, from the aqueous phase (i.e., effluent, supernatant liquor, sludge liquor), incinerated sewage sludge ash from WWTPs or digestates (i.e., digested sewage sludge and manure) from biogas plants, inorganic phosphate fertilizer can be extracted. Numerous technologies have been developed for the various feedstocks. Most of the technologies can be categorized as chemical precipitation, wet-chemical treatment and thermal treatment.

4.1 Chemical precipitation

4.1.1 Principle

Chemical precipitation or crystallization is the simplest way to recover phosphate, and is usually found in the treatment of wastewater stream[68]. Magnesium ammonium phosphate ($MgNH_4PO_4 \cdot 6H_2O$), which is known as struvite may be used as a slow-release fertilizer. Struvite can precipitate spontaneously in municipal WWTPs if the appropriate stoichiometric relations are met and the pH of the solution is slightly alkaline:

$$Mg^{2+} + NH_4^+ + H_nPO_4^{3-n} + 6H_2O$$

$$\rightarrow Mg(NH_4)PO_4 \cdot 6H_2O + nH^+,\ n = 0,\ 1 \text{ or } 2 \qquad (1)$$

The forced precipitation of this substance is the basis of many technologies designed to produce this fertilizer and, when pure enough, is of superior quality to most other recycled phosphate-salts[69].

There are various metal ions in the process liquid such as calcium, aluminum and ferric ions. Therefore, the struvite crystallization is accompanied by the formation of calcium phosphates (which have similar compositions to phosphate rock) or struvite-potassium (K) ($KMgPO_4 \cdot 6H_2O$), where the ammonium in struvite is replaced by K ion)[70].

4.1.2 Application demonstrations

This technology has been extensively researched in laboratory- and pilot-scale operations and adapted to an industrial scale. Especially in European countries, the number of WWTPs with integrated struvite precipitation for sewage treatment is rising. The typical full-scale applications are: OSTARA Crystal Green[71], Crystalactor process[72], REM NUT®[73].

4.1.3 Superiorities and drawbacks

Chemical precipitation/crystallization can extract phosphate from different kinds of wastewaters by adjusting the operating conditions. Phosphate and ammonium can be recovered simultaneously during struvite crystallization.

This can however, also lead to chemical precipitation in pipes, the resulting crystals cause fouling and blockages. Ensuring a certain level of purity of the crystals is difficult because of interfering ions and suspended particles in the process waster[74]. An investment in implementation of this technology is not always economically reasonable, because the costs arising cannot always be covered by the income from recycled phosphate fertilizers, since the phosphate recovery ratio from the aqueous phase is limited. It reaches its maximum in the range of 40%–50% of the WWTP's P load with biological treatment of wastewater[75]. Biological treatment enhances phosphate dissolved in the liquid phase, while chemical treatment retains phosphate in the solid phase.

4.1.4 Influencing factors

Struvite precipitation depends on different parameters and characteristics, such as the pH value, temperature, concentration of interfering ions, fluid dynamics and suspended solids.

4.2 Wet-chemical leaching

4.2.1 Principle

In the wet-chemical method, concentrated phosphate solutions are obtained by washing with suitable chemical solvents. A general concept of wet-chemical leaching is to elute the phosphate from solids by adding of appropriate reagents. According to the use of different reagents, there are normally acidic leaching and a direct alkaline leaching, as described below. It is assumed that most of the phosphates combine with calcium, aluminum and iron ions. The chemical phosphate migrations are presumed to be as follows:

Acid leaching[20]:

$$Ca_9(Al)(PO_4)_7 + 21H^+ \rightarrow 9Ca^{2+} + Al^{3+} + 7H_3PO_4 \quad (2)$$

$$AlPO_4 + 3H^+ \rightarrow Al^{3+} + H_3PO_4 \quad (3)$$

$$Fe_3(PO_4)_2 + 6H^+ \rightarrow 3Fe^{2+} + 2H_3PO_4 \quad (4)$$

$$FePO_4 + 3H^+ \rightarrow Fe^{3+} + H_3PO_4 \quad (5)$$

After acid leaching, phosphate and ammonium are obtained in the liquid phase. Magnesium oxide or magnesium hydroxide is then added as magnesium source. Subsequently, as struvite formation occurs under pH value above 7 (optimal pH 8.5), dosing with an alkaline reagent (i.e., sodium hydroxide) is required to increase the solution pH value.

The phosphates and (heavy) metals ions in the solid phase are released to the liquid phase in an alkaline leaching step[20]:

$$AlPO_4 + 4NaOH \rightarrow 4Na^+ + Al(OH)_4^- + PO_4^{3-} \quad (6)$$

$$3Ca^{2+} + 2PO_4^{3-} \rightarrow Ca_3(PO_4)_2 \quad (7)$$

The amphoteric aluminum bound phosphate then dissolves in the liquid phase and forms calcium phosphate. In the meantime, most of the other (heavy) metals stay in the solid phase. Direct alkaline leaching is less common in comparison to acid leaching. It only produces promising results when used with aluminum-rich and calcium-poor sludge or sludge ash[76].

4.2.2 Application demonstrations

A typical large-scale application of wet-chemical leaching is the Stuttgart process[77], which has been integrated to a municipal WWTP in Germany. This process consists of two major stages: acidic dissolution of metal-bound phosphates, struvite precipitation and crystallization. In this process, phosphate is recovered from sewage sludge in the form of struvite. The SEPHOS process[78] targets sewage sludge ash. Aluminum phosphate is precipitated first by ash acidic elution. A further alkaline treatment can recover calcium phosphate from the precipitate[79]. This is the so-called Advanced Sephos process[78].

Advantages and disadvantages: wet-chemical leaching can be applied to both sewage sludge and sewage sludge ash. With wet-chemical leaching, over 90 wt.% of phosphate in sewage sludge or sewage sludge ash can be eluted.

In the case of acidic leaching, a following liquid-solid separation step (e.g., centrifugation and filtration) is recommended to prepare the liquid phase for the phosphate recovery. The dissolved phosphates and heavy metal ions contained in the liquid phase can then be further processed by separation technologies, e.g., ion exchange, precipitation and extraction[76]. This of course leads to an additional demand concerning chemicals and energy.

Alkaline leaching can extract phosphate directly, however sufficient results are only achieved with certain kinds of waste streams, i.e., with high aluminum, low calcium salt contents. Depending on the local water conditions, this method may be ineffective in some areas.

4.2.3 Influencing factors

The phosphate recovery efficiency via wet-chemical leaching is affected by the acid to ash ratio, as well as by the respective contents of calcium, aluminum and iron counterions.

4.3 Thermal treatment

4.3.1 Principle

There are a lot of thermal treatment possibilities for biomass. Using a hydrothermal process is especially promising for biomasses with high moisture content, since no additional drying step is needed. Depending on the temperature range employed, the hydrothermal processes can be categorized as follows: carbonization[80], liquefaction[81] and gasification[82]. These are all promising technologies to convert biomass into energetic materials and value-added platform chemicals. Solid (hydrochar), liquid (bio-crude oil) and gas (hydrogen or methane, both with carbon dioxide) products can be produced via these three biomass conversion technologies[83].

4.3.2 Application demonstrations

The ASH DEC plant[84] produces phosphate fertilizers from incinerated sewage sludge or manure ash. Through thermochemical treatment, heavy metals are removed from the phosphate-rich biomass. Thus, the bioavailability of phosphate in ash is enhanced.

The AVA cleanphos[85] process was developed to recover phosphate from moist biomass, e.g., sewage sludge or digestate. Followed by hydrothermal carbonization (HTC), phosphate can be recovered from the process product by means of phosphate leaching.

Advantages and disadvantages: considering the relatively high water content of sewage sludge and livestock manure, hydrothermal methods are suitable treatments, due to their ability to deal with wet biomass under moderate reaction conditions, saving time and energy, since no prior drying step is needed[86]. Phosphate extraction from HTC has been highlighted in the past[87,88] due to its moderate reaction condition[89] (temperature around 150–350°C[90], autogenetic pressure) compared to hydrothermal liquefaction (HTL) and hydrothermal gasification (HTG). More than 90 wt.% of the total phosphate contained in hydrochar is in the form of inorganic phosphate salts. Approximately 80 wt.% –90 wt.% of this phosphate can be recovered by subsequent wet-chemical leaching[87]. Since biomass conversion technologies are becoming more and more popular, phosphate recovery via HTL[91] and HTG[92], to obtain value-added chemicals, has been regarded as a promising way to improve the value of these operations.

4.3.3 Influencing factors

The extractable phosphate obtained through hydrothermal treatment obviously depends on the reaction temperature and pressure. Additionally, the pH values of the slurry influence the phosphate transformation.

4.4 Overall assessment

These three phosphate recovery technologies are all well adaptable for use with phosphate-rich biomasses (i.e., sewage sludge, incinerated sewage sludge ash, fresh manure and digested manure) discussed in this review. However, operating conditions have to be adjusted and a combination of the different methods might be required due to the variation in and variety of biomass components.

Among these three technologies, chemical precipitation is the simplest way to extract phosphate from the liquid phase. Still, the overall phosphate recovery ratio is limited by the fact that the liquid phase contains only a small portion of phosphate when compared to the solid phase[57]. Wet-chemical leaching can enhance the migration of the precipitated phosphate from the solid into the liquid phase, this is however accompanied by heavy metal ion dissolution. A combination of these two technologies can concentrate 80 wt.%–90 wt.% of total phosphate in the liquid phase (via wet-chemical leaching), and phosphate can be separated from heavy metal ions and precipitated as bioavailable fertilizer (via chemical precipitation). This process has an excellent phosphate extraction performance on dry biomass, i.e., sewage sludge ash or other dewatered biomasses.

When it comes to phosphate recovery from biomass with relatively high water content, i.e., sewage sludge and livestock manure, either a prior drying or hydrothermal treatment is necessary. These both require intensive energy input, so the economic feasibility is low for commercial utilization[93]. Hydrothermal treatment is normally integrated into biorefinery systems that can convert these agricultural and municipal residues into bioenergy and value-added platform chemicals. The high production cost of a fertilizer with high phosphate (or N) content could be compensated by the production of high-value agricultural products. Phosphate recovery via hydrothermal treatment might be a promising process for the future. A shorter reaction time or milder reaction conditions are beneficial for industrialized application as well.

5 Bioavailability of recovered phosphate fertilizer

P recycling only makes sense if the recovered phosphate can replace the mineral phosphate in the production of fertilizer, which depends on its plant availability. In theory, recovered phosphate should be comparable to mineral phosphate regarding its applicability as fertilizer[94].

Most of the hazardous compounds, i.e., heavy metals and pathogen in sewage and manure are removed, either during the sewage treatment or the intensive phosphate recovery process. There should be no risk of soil contamination and negative impact on crop growth.

Moreover, these recovered fertilizers contain abundant plant nutrients, not only P, N and K, but also other trace elements[95], which are derived from the processed biomass. If their corresponding contents or ratios can be controlled well, these recovered phosphate fertilizers can have an even better performance[96].

Additionally, in the case of HTC we have two products, the phosphate and the hydrochar. There are many studies regarding the potential application of hydrochar as soil amendment[97]. In an optimal scenario the inorganic phosphate fertilizer and the hydrochar would work synergistically to improve the soil quality and increase crop yield.

Due to the diversity of crops and soil types, as well as the varying phosphate fertilizer qualities (depending on biomass feedstocks compositions and treatment methods), only limited tests have been conducted with certain crops, in defined soils with some recovered phosphate fertilizers. Although the assessments carried out are still insufficient, most of the recovered fertilizer trials so far have shown positive results.

Since calcium and magnesium phosphate salts are the most common products obtained from the phosphate recovery processes (calcium and magnesium salt addition during chemical precipitation), most plant availability experiments focus on these two recovered phosphate salts. Struvite, which is a slow-release fertilizer, is superior to calcium phosphates due to its high bioavailability and its applicability to a broader range of soils[96]. Pot experiments have been conducted and verified the positive impact the recovered struvite has on maize cultivation[98].

6 Conclusions

As two representative agricultural production countries, Germany and China are both faced with the impending crisis of the depletion of mineral phosphate resources. However, phosphate-rich waste streams have not yet been considered and recycled to their full potential. A considerable amount of phosphate ends up in sewage sludge and livestock manure. Without a proper management of these residues, the dispersed P can be rarely reused, and causes water as well as soil pollution.

The availability of renewable phosphate in such municipal and agricultural residues was analyzed in relation to their annual production and P contents. Phosphate management relies on the public awareness of the importance of saving environment and resources. There must be certain rules put into place by the legal framework: Germany has strict fertilizer application standards and will enforce phosphates recovery, while China is more inclined toward the use of waste resources than recycling. Along with the implementation of definite phosphate recovery regulations Germany is trying to close the P cycle. An overview of different phosphate recycling concepts was given in this review. It has been a long-term effort for Germany to push the phosphate recycling into practice along with applying innovative phosphate recovery technologies into full-scale operation.

Regarding the differences in agricultural infrastructure, population base, agricultural residues amounts and ratios, it is not feasible to apply the German experiences of phosphate recycling directly to China. China has to develop its own phosphate recycling strategy tailored to its own characteristics and needs. It is beneficial for China to selectively learn from the experiences of the establishment of phosphate cycle in Germany. This also constitutes a technological leapfrogging between Germany and China: the global P crisis is a challenge for both countries, but it also provides the opportunity for a cooperation to develop more cost- and energy-effective phosphate recovery technologies and thereby close the P cycle.

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