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STUDIES AND RESEARCH ON PROCESSES FOR THE ELIMINATION OF CADMIUM FROM PHOSPHORIC ACID*

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RESUME

Avec l'accord de l'IMPHOS qui a commandité cette étude et de la Commission des Communautés Européennes qui l'a financée, je propose d'en présenter l'essentiel au Congrès Technique de 1994.

Après exposé de la position de l'industrie des engrais vis-à-vis de la problématique du cadmium dans les engrais (USA/Europe/Afrique), on fera la synthèse des 4 pistes de recherche principales, à savoir :

- *la co-cristallisation du cadmium dans l'anhydrite :* *procédés CC*
- *la précipitation du Cd par des molécules contenant S^{--} :* *procédés PP*
- *l'enlèvement du Cd par résines échangeuses d'ions :* *procédés RX*
- *l'enlèvement du Cd par extraction aux solvants :* *procédés SX*

Le cadre étant ainsi tracé, on évaluera les chances de succès et les coûts des différentes filières (sans référence directe aux procédés particuliers qui les mettent en oeuvre) et on situera le degré d'avancement des travaux.

On passera ensuite à l'examen critique des filières, du sort des rejets et on examinera le ou les contextes où pourrait se faire la décadmiation en fonction des spécificités des filières. On terminera par la question du cadmium dans les engrais en fonction des éléments connus.

Des éléments de conclusion seront alors exposés.

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1. FOREWORD

Following P. Becker's 1988-89 report entitled "Inventory of Studies and Processes concerned with the elimination of Cadmium from Phosphoric Acid" to the EC Commission, I have been asked, as a result of an agreement in 1991, to update and extend this study.

This mission was financed by the EC Commission, at IMPHOS' request; it involved contacts with about 30 companies concerned in West Europe, Africa and USA.

I want to thank IMPHOS and D.G. VIII of the EC Commission for the permission to present a summary of the results of that mission to this IFA Technical Conference.

2. ASPECTS OF THE PROBLEM

As could be expected, Sweden, known for its proselytism in that field, first drew attention on the presence of cadmium in phosphate fertilizers during the 80's.

At the request of EC Commission, the European Fertilizer Manufacturers' Association (EFMA) summed up the situation as follows in 1989.

For the whole of EEC, the average cadmium content of fertilizers is 60 mg/kg P_2O_5 which means that, if the distribution of fertilizers was uniform, the amount of cadmium applied would be 2.5 g/ha/year or less than 1 microgram cadmium per kg of cultivated soil. Knowing that the average cadmium content of soils is 0.5 mg per kg, 500 years would be needed to double the soil content if no cadmium was taken up from the soil!

*Original article in French

But plants absorb cadmium according to availability which depends on the soil type, its characteristics, in particular the pH and the presence of other metals. A fraction of this cadmium enters the plants and, hence, the food chain, either directly, or through animals, so that to-date, there is no proven evidence of a relationship between soil cadmium and human health.

In addition, phosphate fertilizers are not the only source of cadmium in the soil. There are at least three other sources, namely:

- rains, where a range of 1 and 8 g/ha with an average of 2 to 4 g/ha are cited.
- sewage sludge spread on the soil for which the 1986 European Directive mentioned a limit of 150 g/ha/year as a ten year average
- slurry applications for which the average figures are not available, but which may raise serious problems in intensive livestock areas.

These three sources together gave an annual amount of 350-400 t cadmium in the 1980's while fertilizers supplied 250 t cadmium annually.

Considering that there was no satisfactory technology to reduce the cadmium content of fertilizers, EFMA suggested to limit as much as possible any restriction to the supply of cadmium by fertilizers only to areas felt critical and to the assessment of the contribution of fertilizers in the increase of cadmium content of these soils. In fact, the limitation of Cd supply would mean excluding the use of phosphates coming from traditional suppliers **(Appendix 1 and 2)**.

3. POSITION OF FERTILIZER PRODUCERS

As could be expected, fertilizer producers respond according to the constraints they undergo.

In the US, the cadmium content in fertilizers does not raise any problem and fertilizer producers are not concerned with the problem, even more so as Florida phosphate rock, the main export basis, has a low cadmium content. However, research centres took interest in processes able to remove magnesium from phosphoric acid and it appeared that cadmium was separated in the same operation.

In Europe, the constraint is heavy. Limits in cadmium content is legally binding in Scandinavia, Switzerland and Japan, contractual in Germany and Belgium, pending in other countries and as a result, producers react:

- producers of fertilizers by the nitric acid route cannot remove cadmium; they then moved towards low cadmium phosphates
- Those who use phosphoric acid have difficult times; they have been decimated by outside competition and the supposedly internal ecological constraints. Their efforts to control the cadmium content of phosphoric acid often lead them to so expensive processes that are not suitable for fertilizer production. Many have abandoned their research on decadmiation and have also chosen low cadmium phosphates. That situation makes it impossible to export to Europe phosphates with high cadmium content and fertilizers made out of them.

For the African producers, that constraint is so detrimental to their supply of phosphate rocks and intermediates to their natural market, Europe, that these phosphate producers are seriously involved in numerous investigations, either by themselves, or in co-operation with research centres.

In addition, the European Community, feeling somewhat guilty of the initiatives of some of its members depriving countries like Senegal and Togo, maybe later Tunisia, of their resources, took part in financing some of these researches, and were joined by the World Bank, to a lesser extent.

It is not surprising that IMPHOS, an organization set to promote and protect phosphate producers in Africa and the Middle East took initiative in asking EEC to finance a mission to study the position of that question.

4. PRESENTATION OF THE MAIN RESEARCH LINES

Successive individual contacts gave me the impression of an infinite variety of research tracks; and since each company or organization considered the problem in its own way, there seemed to be as many tracks as companies. But convergence of reasonings made it possible to put together all the researches which were developed independently under various conditions to be classified in the following four groups:

PRINCIPLE OF THE PROCESS	IDENTIFICATION OF THE ROUTE
1. Co-crystallization of cadmium with anhydrite	CC
2. Precipitation of cadmium by molecules containing sulphide ion	PP
3. Removal of cadmium by an ion exchange	RX
4. Removal of cadmium by solvent extraction	SX

5. CHARACTERISTICS OF THE ROUTES AND STATE OF PROGRESS

5.1. CC Route

- Includes processes which induce the precipitation of calcium sulphate as anhydrite, which means operating at high temperature and high acidity (phosphoric and sulphuric acid together).

- Anhydrite promotes the co-crystallization of dicalcium phosphate, cadmium and some rare earths.

- That category includes two processes which cause the precipitation of anhydrite in a different way:

. The OTP-PB process (Office Togolais des Phosphates - Pierre Becker) proceeds with a post-treatment of the merchant acid.

. The OCP process (Office Chérifien des Phosphates) adjusts the conditions of the concentration to induce the precipitation of anhydrite.

- State of progress in 1992 - Laboratory pilot scale.

- Possible decadmiation efficiency: more than 90%.

- Wholly mineral route, using phosphate rock and sulphuric acid to produce calcium sulphate, e.g. all products existing in the phosphoric acid plant.

- Has to be operated on a phosphoric acid production site.

- Residues: about 70 kg/ton P_2O_5 containing 0.1-0.3% Cd.

- The post-treatment of these residues is possible to separate cadmium and rare earths.

- Intrinsic added value: produces a desaturated merchant acid, with no post-precipitation.

- Possible added value: possibility of selling cadmium and rare earths concentrates after post-treatment of the residues.

5.2. PP Route

- Includes processes which precipitate cadmium by the reaction with sulphide ions generally in the form of phosphoric acid dithioester of dialkyl reacting with the filter acid or the merchant acid.

- This category includes four processes differing in operational details such as the choice of the precipitant, its action mode and, especially, the method of separation of the precipitate.

- The process developed by Hoechst uses reagents produced by the firm in proportions equal to 8-10 times the stoichiometry to precipitate more than 90% of the cadmium; the separation is achieved by using additives; the residues contain 1% Cd. The laboratory pilot scale stage is over. The process is said to be ready for industrial use (via Uhde). Research has stopped.

- The process developed by SIAPE is similar in its principles but different as the additives come from various suppliers and are used in limited amounts to control the cost while accepting that a decadmiation limit of 75% usually sufficient. In addition the acid is pre-treated and the filtration additives also include gypsum. The process is applied in practice in a unit processing 100,000 t P_2O_5 per annum for use as cattle feed.

- The URANIUM PECHINEY - IRCHA - CRVM process includes the following variants:
  . It requires the preliminary reduction of the acid
  . It uses sodium ester salt
  . The separation is achieved by ionic flotation.

The process was tested in laboratory pilot plant on synthetic acids and one sample of Togo acid. Research has stopped.

- The OCP process involves the following variants:
  . Pre-treatment of the acid (from filtration)
  . Precipitation by dialkyl dithioesters or H_2S, Na_2S, NaHS, even P_4S_{10}
  . Separation of the precipitate by acetone treatment followed by filtration of the precipitate and distillation of the filtrate to separate acetone, which is recycled.

The process was tested in laboratory pilot plant on Moroccan acids. Research has stopped.

- Route using a costly organic precipitating agent, but gave proven results, the cost of which, unacceptable for fertilizers, could be considered for more valuable uses of phosphoric acid.

- Can be disconnected from phosphoric acid manufacture.

- Residues: can be concentrated to 1% Cd (and even 18% Cd in the last two processes mentioned) and, as a result, are not very abundant.

- Could be treated to give a source of cadmium.

- Intrinsic added value: removes also Zn, Cu, As, Mo, Zr and possibly U.

5.3. RX Route

- Includes processes which separate cadmium by the action of ion exchange resins; two of the four processes of that route were developed in Florida to separate magnesium which shows an increasing concentration as rock mining moves further South.

- These processes vary according to the type of resin, and the mode of physical and chemical operation.

- PECO process is characterized by the extraction of divalent ions by cationic resins: Mg^{++}, Ca^{++}, Cd^{++}, Fe^{++} using resin in fixed bed and regeneration by a solution of ammonium sulphate.

- PEMBROKE cationic resin process extracts the same divalent ions using a system of fluidized bed of resins and regeneration by a solution of ammonium sulphate.

- The ENICHEM process uses resins successively mixed then anionic to extract cadmium bromide using a system of fixed or fluidized beds and recycling of the eluted solution to reach 15% Cd.

- The process developed by HYDRO AGRI ROTTERDAM, which will be presented by its authors at this IFA Technical Conference, uses anionic resins to extract a cadmium halogenide.

- It is generally agreed that phosphoric acid must follow a desaturation pretreatment before getting in touch with the resins although promoters often refuse to admit it.

- State of progress: laboratory pilot scale on settled acid

- Possible rate of decadmiation: can reach 90%.

- Residues: variable for different processes; ENICHEM only studied the premium product uses: by precipitation as CdS and centrifugation, it produces a feedstock to the cadmium industry.

- Must be close to the phosphoric acid plant

- Added values: also remove other divalent ions. The pretreatment can be advantageous to the phosphoric concentration unit.

5.4. SX Route

- Includes a process extracting cadmium by using a solvent made of a salt of tertiary amine diluted in an aliphatic hydrocarbon.

- The re-extraction of cadmium is then achieved by an aqueous phase then submitted to chemical and thermal treatments to isolate cadmium and concentrate it in a basic residue.

- State of progress: BUDENHEIM started a 200 t/d P_2O_5 unit in 1992.

- Rate of decadmiation: more than 90%.

- Requires a pretreated acid to obtain a complete desaturation, which makes it too costly for acid used to produce fertilizers, but the cost is bearable for acid used in a more profitable way.

- Residues: contain 15 to 20% Cd and after neutralization with lime. Can be put in barrels and stored as toxic material in salt mines.

- Can be established outside phosphoric acid production site.
- Added values: remove the other minerals; the pre-treatment can be beneficial for further operations.

6. COMPARISON OF THE ROUTES UNDER CONSIDERATION

6.1. Specific aspects of the routes

CC route must be specifically adjusted to the phosphate rock processed; indeed, as in the case of phosphoric acid manufacture, it calls on calcium sulphate crystallization, which is influenced by the various impurities present in the phosphate treated. As the impurities are different from one rock to the other, operational conditions must be adjusted to each case. This appeared in the tests with the OTP/PB process and as well as in the OCP process.

PP route which uses a cadmium-specific precipitating agent does not depend upon the phosphate processed except for the viscosity of the acid which influences the separation of the cadmium precipitate (flocculation or not, duration of the operation, carry over of P_2O_5 into the precipitate).

RX and SX routes are less dependent on the phosphate treated since the acid pre-treatment equalizes the quality to some extent.

6.2. Economic comparisons

To evaluate the relative and absolute interest of the various routes mentioned, it is of paramount importance to evaluate the cost under standardized conditions; the assumption retained for that purpose is that of 500 t/d P_2O_5 plant located in West Europe.

The costs of route CC were calculated with the assistance of an engineering office using data supplied by the promoters of the processes. Since it refers to operations very similar to those met in phosphoric acid plants, thus very familiar to all parties, one can state that they correspond fairly well to the reality.

The costs for PP route were calculated in the same way and the same remarks apply.

Regarding RX and SX routes, my personal experience as well as the opinion of specialists make me consider only variants "with acid pre-treatment", the estimated costs of which appear to be the minimum necessary to make these processes operational.

6.3. Critical appraisal of the routes studied

CC is the route which can be best included in phosphoric acid production: it uses the same equipment and the same raw materials.

It offers an important assured added value: the production of an acid with no tendency of post precipitation, ensuring economies in storage and transport.

Another possible added value, consequently uncertain: the potential of the metal content: rare earths and cadmium.

PP route is applicable to merchant acid as such (although some promoters prefer treatment before concentration).

SUMMARY

SUMMARY OF INVESTMENT AND OPERATIONAL COSTS OF VARIOUS PROCESSES FOR DECADMIATION OF PHOSPHORIC ACID IN A 500 T/D P_2O_5 UNIT LOCATED IN WEST EUROPE						
PROCESSES			INVESTMENT (million)		COST (per t P_2O_5)	
Type	Conditions		USD	ECU	USD	ECU
CC	Without residue treatment	Without valorization	3.30	2.70	8.65	7.00
		With valorization			5.65	4.50
	With residue treatment	Without valorization	4.35	3.50	10.10	8.15
		With valorization			5.65	4.50
PP			4.25	3.40	20.00	16.00
RX	With medium acid pretreatment		6.20	5.00	25.00	20.00
SX	With full acid pretreatment		6.30	5.10	27.00	21.75

IMPORTANT REMARKS:

1. At this stage of process development, these costs have only a relative and indicative value since they may increase substantially as a result for example of substantial P_2O_5 losses or difficulties in the residue treatments.
2. To estimate them, they must be compared to the market price of phosphoric acid, which fluctuates around US$ 300 per ton P_2O_5. Decadmiation would in the best case represent only 2 to 3% of the selling price of the acid.
3. The cost of decadmiation could be reduced even more if the objective would be reached by mixing decadmiated acid and crude acid; in that case, the reduction would be proportional to the rate of incorporation of crude acid.

- It is more independent of the production of phosphoric acid, but some versions prefer proximity to production site.
- In some versions presented, there is an added value of acid quality.

RX route, according to phosphoric acid specialist's opinion, will require a pretreatment of the acid (which can be regarded similar to the one achieved when using CC route).

- produces a purified acid liable to take part in usages more demanding than fertilizer manufacture.

SX route will request a more thorough acid pretreatment than the previous one (thus more costly than that obtained by the CC route).

- It also produces a purified acid which can accommodate more rigid specifications than those required by fertilizer manufacture.

7. THE RESIDUE PROBLEM

Removing cadmium from phosphoric acid - at least partially - is one thing, but its discharge in a concentrated and available form must be avoided.

The best solution would be to isolate the cadmium concentrate which could be supplied as feedstock to the cadmium industry.

But, under the pressure against the use of cadmium, the market of that metal and its derivatives is in a prolonged slump. Inevitably, they are already covered by the by-products of the zinc industry. Since the cadmium industry is not to be expected to absorb all the amounts of the phosphate industry, therefore part of it at least should be treated as an industrial waste.

Let us consider the situation of the various routes:

CC route:
- Bulky residue amounting to ± 70 kg/t P_2O_5, but with a low cadmium concentration (less than 1%)
- It can be neutralized and stored on a watertight ground
- As an alternative, it could be treated to recover rare earths and a cadmium concentrate (25-30% cadmium) which can be neutralized and stored as toxic waste.

PP route:
- Bulky residue to be treated as above for some versions
- More concentrated residue in other versions (up to 1-2% Cd) which can be deactivated and stored on a watertight ground or treated to extract a cadmium concentrate.

RX and SX routes:
- Liquid residues very low in cadmium to be treated - for example with sulphide - to extract a cadmium residue, as above.

In general, the post-treatment of residues has not been extensively nor very thoroughly considered. Under those conditions, it seems premature to use it as a criterion for the comparison of the processes.

8. CONDITIONS IN WHICH DECADMIATION COULD BE IMPLEMENTED

Depending on circumstances, this operation can be applied:

- **either in the chemical complex where phosphoric acid is produced**

In that case, all the routes can be used, but CC route, if its industrial upscaling is successful, would be the most economical.

PP route could be competitive if the price of reagent would decrease, either with lower selling prices, or through recycling.

Under the present state of knowledge, RX and SX routes have no possibility since they are too costly because of the required acid pre-treatment.

- **or in an intermediate storage or at a big consumer.**

CC route is excluded since it requires a link with a phosphoric acid plant.

PP route can be adapted but the treatment of residues is necessary and increases costs.

RX and SX routes are not more competitive than in the previous case and would possibly be justified only in association with a non-fertilizer outlet for the acid.

9. REAPPRAISAL OF THE CADMIUM PROBLEM IN FERTILIZERS

The toxicity of cadmium supplied by phosphate fertilizers is not proven. Indeed, its uptake by plants depends on their bio-availability which itself varies with different factors, of which the main one seems to be the soil pH. Some experiments showed that, by increasing the soil pH from 5.6 to 6.6, the availability of Cd is reduced six fold. The interaction with other metals also reduces the availability significantly.

Then, one should notice that all the cadmium absorbed by the body is not metabolized; actually, 95% of the cadmium is excreted through the natural channels.

As a result, the problem should not be over-emphasized, in particular:

- one should remember that there is very little cadmium moving from the plants to human beings.

- that there is no very simple way of removing cadmium from phosphate fertilizers.

The situation can be summarized as follows:

  . the processes proven at the industrial scale are not economical in the case of acid used for fertilizers

  . the processes which are economically acceptable for fertilizer purposes are still at the laboratory pilot stage and the most difficult part of the development has still to be made requiring time and money.

- accept that there may be more hazardous local situations, in particular:
  . where the soils are the most acid (Scandinavia for example).
  . where sewage sludge is applied in large quantities
  . where the livestock population has reached a non-sustainable level, since the land available is not sufficient to accommodate the slurry produced which means that the regions concerned must export it with its inherent nuisances either as such or after-treatment
  . where the emission to the atmosphere is significant - near factories processing cadmium containing materials.

The studies made by:

- A.P.A.O. entitled "Phosphates and cadmium: soil, plants, men"

- The Plant Biology Department of the Agronomic Faculty of Gembloux entitled "Relative importance of phosphate fertilizers in the contamination of the environment by cadmium", agrees that, in the present state of knowledge and at the amounts encountered in normal living conditions, no "accident" is likely to occur through food ingestion. This does not exclude to caution in "hazardous areas" as defined above but should dismiss any idea of an improvised general legislation concerning the contribution of phosphate fertilizers as the source of cadmium, since the so-called remedy could be worse than the problem. The result would be a complication of the operations which would be more energy demanding, involve more complicated handling and give more toxic cadmium residues, while phosphate fertilizers maintain about the same content in fertilizers as in rock phosphates.

In addition, the unavoidable increased cost of fertilizers resulting from such a legislation would be at the expense of developing countries where the use of fertilizers should increase in order to avoid more serious human problems.

10. CONCLUSIONS

Allow me to present a conclusion in the form of a syllogism.

10.1. Key Issue

When some people are concerned with the cadmium supplied by phosphate fertilizers and propose to limit it drastically, they do not deal with the actual problem since they confuse total and available cadmium.

Indeed, in the phosphate, then in fertilizers and finally in the soil, the total amount of cadmium represents only a biological abstraction, since the rates of **bio-availability** vary enormously; but only these rates influence the uptake by the plant, prelude to the absorption by human beings. The cadmium existing in phosphates, fertilizers or the soil is never alone: it is associated with other metals and one knows that multi-metal interactions are essential for the substitutions, the circulation of fluids and the translocation in the plant; the inhibiting role of iron, among others, is well known by specialists (studies conducted by INRA and CIRAD under APAO guidance).

In addition, an epidemiological study conducted in France by C. Boudene showed that the standard diet does not noticeably vary with time, nor in the space and that its cadmium supply is about one third of the amount which a man can absorb safely according to W.H.O.

10.2. Secondary Issues

In addition, in the last ten years, the total amount of cadmium released has decreased substantially.

First, the atmospheric contribution of cadmium decreased strongly as a result of:

- the crisis which induced reduction in industrial production
- the relocation of companies
- the new developments in the purification of industrial wastes

On the other hand, the fertilizer consumption decreased for three reasons:

- the Common Agricultural Policy (CAP) of the European Union which imposes fallowing of noticeable proportion of arable land,

- the development of intensive livestock feedlots using imported feedstuffs and producing residues which supply the soil with additional nutrients, thus reducing the mineral fertilizer requirements.

- the intrinsic decrease of fertilizer use as, responding to the criticism of pollution due to excess fertilization, the agricultural community developed the idea of controlled fertilizer application to ensure a sustainable soil fertility. Under these circumstances, the total amount applied should not exceed the removals except in cases of obvious imbalances.

10.3. Final suggestion

Summarizing, we can remember that we are not in a state of serious crisis. If there were a hazard, it would be in the very long term. It seems obvious to me that, in our difficult world, everyone should keep calm and avoid needless instability in these fragile economic equilibria. Under the circumstances, to tackle seriously the problem of cadmium supply by fertilizers, I propose a 10 year moratorium and then to take advantage of that period to study that question carefully and in depth by:

- Studying the relative importance of phosphate fertilizers in the contamination of plants and the risks that the latter present to human beings.

- Promoting the development of the most promising process(es) of decadmiation of phosphoric acid performance-wise, namely those of the CC route, possibly PP route if there was a good project. But these processes are only at the laboratory stage and they must undergo the phase of industrial pilot plant where costs are high and income almost nil, since the quantities produced are used to test the market, not to cover it.

10.4. Final reflection

If the proposed studies are seriously carried out, in about 10 years, we will have an objective appraisal of the possible hazards and, if it is felt necessary to reduce, in some areas and situations, the cadmium supply by fertilizers, one can hope to have a phosphoric acid decadmiation technique of a bearable cost. Thus we would not or no longer exclude from the market phosphates known and sold for decades.

And, as I stated in my report to the EEC: "A rigorous scientific reasoning and honest logical deductions could substitute the hypocrisy of those who associate the romanticism of gratuitous assumptions and the pretension of hazardous inferences".

Doing so, we would be quite in line with the "Heidelberg Call"[1].

[1] The solemn call was launched during the United Nations Conference on Environment and Development, held in Rio de Janeiro, in June 1992. It came from the 264 signatories (including 52 Nobel Prizes winners) and which is known under "Heidelberg Call". They warned against any "irrational ideology which opposes scientific and industrial progress and is detrimental to economic and social development".
The permanent secretariat of the "Heidelberg Call", based in Paris, received the signatures of outstanding scientists (more than 2700 signatories representing 102 countries and 72 Nobel Prizes winners) who commit themselves to use correct scientific methods in the treatment of environmenal questions in the whole world.

APPENDIX 1

AVERAGE CONTENTS IN P_2O_5 AND Cd OF THE MAIN COMMERCIAL PHOSPHATE ROCKS

MERCHANT PHOSPHATES	P_2O_5 % total	CADMIUM ppm / total	ppm / P_2O_5	ppm / P
1. IGNEOUS ORIGIN				
Kola	39	< 5	< 13	< 30
Pharlaborwa	37	< 5	< 13	< 30
2. SEDIMENTARY ORIGIN				
Florida	32	7.5	23	54
Jordan	33	< 10	< 30	< 70
Khouribga	32/33	15	46	106
Syria	31	16	52	119
Algeria	29	17.5	60	138
Egypt	27	20	74	170
Bu-Cra	34	34	100	229
Nahal Zin	31	31	100	229
Youssoufia	33	40	121	277
Gafsa	29	40	137	315
Togo	37	60	162	371
North Carolina	30	50	166	381
Taiba	37	75	203	464

APPENDIX 2

PERMITTED RATE AND PHOSPHATES EXLUSION

PERMITTED RATE mg Cd / kg P_2O_5	PHOSPHATES EXCLUDED
250	None
200	Senegal
150	Senegal, Togo, North Carolina
100	Senegal, Togo, North Carolina, Tunisia, Morocco (Youssoufia), Israel (partially)
50	Senegal, Togo, North Carolina, Tunisia, Morocco (Youssoufia), Morocco (Bu-Cra), Israel (totally), Algeria, Egypt, Syria