Effect of Long-Term MSW compost Applications on Metal Uptake by Cucumber (Cucumis sativus 1.) Plant and Soil metal Bioavailability

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Abstract— A greenhouse experiment was carried out to assess heavy metal loading and its bioavailability on the greenhouse soil and uptake of metals by Cucumber plant following MSW (Municipal solid waste) compost application. MSW compost was applied from 2005 to 2009 to greenhouse soil as an oven-dry basis, totaly at 100 ton ha⁻¹(MSWC₁₀₀) and 200 ton ha⁻¹ (MSWC₂₀₀) levels. An uncontaminated greenhouse soil at the same location was used as a control treatment for comparison with the results. After the end of 4 years of successive MSW compost applications to greenhouse soil, the residual effects of MSW compost on heavy metal accumulation in cucumber and total and bioavailable heavy metal status of greenhouse soil were tested.

It was found that plant heavy metal contents were higher in MSW compost treated greenhouse soil than that in untreated soil. Higher levels of MSW compost applications caused higher accumulation of Zn, Cu, Ni, Pb and Cd in the leaves and fruits of cucumber plant. Concentrations of heavy metals in plants were below the phytotoxic levels. In the MSW compost treated greenhouse soil, according to background and toxicity limits, heavy metal status of Cucumber leaves was ranged in normal and high levels. Although concentrations of heavy metals in cucumber fruit were below the phytotoxic threshold levels, Pb concentration was exceeded safety food metal limit value for edible vegetables by MSWC₂₀₀ compost treatments.

The greenhouse soil treated with MSW compost contained higher concentrations of both total and DTPA-extractable Zn, Cu, Ni, Pb and Cd than that of untreated soil. Results obtained for DTPA extractable metal availability indicated higher relative metal availability in MSW treated soils. Relative metal availability order of metals in MSW treated soils were Zn>Pb>Cu>Cd>Ni. Bioavailable Cd content of control soil was found below the detection limit. The amount of bioavailable metals in the greenhouse soil was significantly high for MSW compost treatments. In MSW compost treatments, total soil metal concentrations were found below the permissible pollutant limits, but the increase in available metal fractions was more marked than those of total concentrations.

Keywords— MSW compost, heavy metal accumulation, metal bioavailability, cucumber.

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I. INTRODUCTION

Composting of municipal solid waste (MSW) and its subsequent application to agricultural land is gaining popularity because of environmental concerns associated with the disposal of this material in landfills. Several studies have shown that use of MSW compost in agriculture has many benefits to soil, crops and environment [1].

However, MSW compost often contains potentially toxic elements, that can cause soil contamination, phytotoxicity and undesirable residues in plant and animal products [2]. As a matter of fact, pollution problems may arise if toxic metals are mobilized into the soil solution and are either taken up by plants or transported in drainage waters. Risk for human health may then occur through consumption of such crops and intake of contaminated waters. In the long term, the use of MSW compost can also cause a significant accumulation of Zn, Cu, Pb, Ni and Cd in the soil and plants [3].

The maximum permissible concentrations of heavy metals in surface soils amended with sewage sludge or MSW compost are normally based on total concentration, although it is the bioavailable metal fraction that posses environmental concern [4]. Nevertheless, these criteria are insufficient since mobility, environmental diffusion and bioavailability largely depend on soil physico-chemical characteristics and, likewise, on trace metal chemical forms [5]. From an environmental point of view, the evaluation and forecast of food contamination is related to the bioavailable fraction of heavy metals in soil.

Information on the fertilizing value of MSW composts and their effects on the heavy metal loading potentials on greenhouse soil are scarce. The aim of this study was to assess the residual effects of successive MSW compost applications on the total and bioavailable (EDTA-extractable) contents of Zn, Cu, Ni, Pb and Cd in the greenhouse soil, and heavy metal accumulation in Cucumber leaves and fruits.

II. MATERIAL AND METHODS

The experiment was conducted from 2005 to 2009 on the greenhouse representative of the major greenhouse vegetable growing area of Turkey Antalya Aksu. The analytical characteristics of the greenhouse soil and MSW compost are shown in Table 1 which also shows the pollutant limits of soil

and also organic materials used as soil amendments, permitted by EU legislation [6].

| TABLE I. THE ANALYTICAL CHARACTERISTICS OF THE |
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| EXPERIMENTAL SOIL AND MSW COMPOST BEFORE |
| TREATMENT, AND THEIR POLLUTANT LIMITS. |

| Parameters | Soil | | Limit | MSW | Limit values in | | | | |
|---------------------------|-----------------|----------|----------------------|-------------------|------------------------|--|--|--|--|
| | | | values in | compost | organic | | | | |
| | | | soil [6] | | materials[6] | | | | |
| Texture | Loam | | | - | | | | | |
| pH- H ₂ O (1:5 | 7.37 | | | 7.55 | | | | | |
| w/v) | | | | | | | | | |
| CaCO ₃ , % | 7.68 | | | - | | | | | |
| Total N, % | 0.17 | | | 0.68 | | | | | |
| Organic | 2.33 | | | 58 | | | | | |
| Matter, % | | | | | | | | | |
| EC ($dS m^{-1}$) | 0.04 | | | 8.77 | | | | | |
| Zn, mg kg ⁻¹ | 77 ¹ | 14^{2} | 150-300 ¹ | 1110 ¹ | 2500-4000 ¹ | | | | |
| Cu, mg kg ⁻¹ | 17 | 5 | 50-140 | 96 | 1000-1750 | | | | |
| Ni, mg kg ⁻¹ | 14 | 0.5 | 30-75 | 47 | 300-400 | | | | |
| | | 5 | | | | | | | |
| Pb, mg kg ⁻¹ | 33 | 8 | 50-300 | 164 | 750-1200 | | | | |
| Cd, mg kg ⁻¹ | * | * | 1-3 | 1.3 | 20-40 | | | | |

*: Below detection limit (< 0.02 mg kg⁻¹), ¹: Total concentrations (mg kg⁻¹ dry wt), ²: DTPA-extractable concentrations (mg kg⁻¹ dry wt)

MSW compost was obtained from MSW composting plant in Kemer, Antalya. Compost was produced by the composting of the organic fraction of unseparated municipal solid waste, selected mechanically at the plant. MSW compost was applied from 2005 to 2009 to greenhouse soil as an oven-dry basis, totaly at 100 ton ha^{-1} (MSWC₁₀₀) and 200 ton ha^{-1} (MSWC₂₀₀) levels. In the period of February 2005-2009, 25 % of the total amount of MSW compost to be used for each treatment was distributed. MSW compost was manually incorporated into the greenhouse soil and mixed throughout the upper 20 cm. An uncontaminated soil for the control treatment was used to compare the effectiveness of MSW compost application in the same greenhouse with all agronomic practices except MSW compost application.

The experimental plots have been cultivated in four consecutive years 2005-2009 with Cucumber (*Cucumis sativus* L.) at a density of 38200 plants ha⁻¹ and each plot (5.25 m^2) consisted of 20 Cucumber plants in a randomized block design with four replications. The complete greenhouse operations were carried out in the trials. Basic fertilizer was applied with sprinkler irrigation in all plots at the rate of 300, 200 and 300 kg ha⁻¹ of N, P and K, respectively.

During the experiment, the plants were watered regularly and treated according to common agrotechnical principles. For the determination of heavy metals, both plant and soil samples were taken at the end of experiment, 2009. At the beginning of the flowering period leaf samples were collected from each treatment, and fruits were sampled at the full ripening stage. After the harvest of cucumber, soil samples were taken at a depth of 10-20 cm in September 2004; and these were airdried and sieved (< 2 mm). Leaves and fruits of cucumber were analysed separately. Plant samples were dried at 60 °C in a forced-air oven, ground and then digested in aqua regia (1:3 $HNO_3/HCl)$. For the determination of 'total' heavy metal concentrations, soil and MSW compost samples were digested in aqua regia (1:3 HNO₃/HCl) according to the international standard [7]. Bioavailable fractions of metals were extracted from soil with diethylenetriaminepentaacetic acid-CaCl₂-triethanolamine adjusted to pH 7.3 (DTPA) [8] procedure. Total and bioavailable Zn, Cu, Ni, Pb and Cd concentrations of greenhouse soil and plant metal content were determined by flame atomic absorption spectrometry (FAAS) under optimised measurement conditions, and values were adjusted for oven dried (12 h at 105 °C) material.

A statistical ANOVA F test was applied to the results and, treatment means were compared by the least significant difference test at P<0.05.

III. RESULTS AND DISCUSSION

Plant Heavy Metal Contents

Concentrations of Zn, Cu, Ni, Pb and Cd in the control treatment were small and representative of background levels [9] (Table 2). The concentration of Cd in the fruits of Cucumber plant grown in control medium was small and below the detection limit of analytical apparatus. As would be expected, heavy metal concentrations of Cucumber leaves in MSW compost treatments were higher than that of control treatment. Higher levels of MSW compost applications caused higher accumulation of Zn, Cu, Ni, Pb and Cd in the leaves and fruits of cucumber plant.

Limit values of Pb and Cd in edible vegetables were suggested as 0.25 and 0.1 mg kg⁻¹ in fresh material [10] (corresponding about 2.5 and 1.0 mg kg⁻¹ dry weight basis, respectively). Although Zn, Ni, Pb and Cd contents of cucumber leaves in MSW compost treatments showed that the amounts were significantly below the phytotoxic threshold levels [11], foodstuff index and limit values of Pb and Cd in Cucumber fruit were exceeded by MSW_{200} compost treatments (Table 2).

Total metal contents in the soil

The heavy metal contents of untreated greenhouse soil and organic materials studied (Table 1) are well within the accepted normal range of values. A comparison of metal contents of organic materials with that of untreated soil showed that the metals Zn, Cu, Ni, Pb and Cd were present in MSW compost in greater concentrations than in the soil. The heavy metal concentrations of MSW compost is below the levels indicated by the EU [6] for the agricultural use of waste organic material (sewage sludge).

Successive applications of MSW compost for 4-yr period led to a far greater introduction of the heavy metals examined and brought about a significant increase in their 'total' form in the soil when compared with the control (Table 3). After 4 years of successive MSW compost applications, significant changes in the total contents of Zn, Cu, Ni, Pb and Cd among the treatments were determined. These results agree with the increase of total metal content observed in soils submitted to repeated application of metal-contaminated sewage sludges [12]. The total Zn, Cu, Pb and Ni contents of soil at the beginning of the experiment were 77, 17, 33 and 14 mg kg⁻¹, respectively. Total Cd concentration in the uncontaminated soil was below the detection limit of analytical apparatus ($<0.02 \text{ mg kg}^{-1}$). During the trial period, at high application levels of MSW compost, these levels were increased to 161, 28, 59 and 23 mg kg⁻¹, respectively, and also Cd level was 0.16 mg kg⁻¹. The Zn, Cu, Pb and Cd variations were more marked with sewage sludge and consistent with the amounts of MSW compost tested. Increasing soil-metal concentrations with increasing MSW compost applications have been reported [13, 14]. In spite of the important increases in Zn, Cu and Pb contents registered, the concentrations in the soil remained below the EU legislation [6] for soils.

DTPA extractable metals in the soil

All amounts of MSW compost brought about significant increases in DTPA-extractable metal concentrations in comparison with the control (Table 3). DTPA-extractable Zn, Cu, Pb, Ni and Cd also registered significantly higher values in 200 ton ha⁻¹ than in 100 ton ha⁻¹ application level of MSW compost. DTPA extractable Cd concentrations of control treatment were always below the sensitivity of analytical method (0.02 mg kg⁻¹).

To compare the relative availability of different soil metals, DTPA/Total ratio is used to give available metals as a percentage of total soil metal. The relative increases of Zn, Cu, Pb, Ni and Cd for MSW compost in comparison with the control were higher for the DTPA-extractable than for the 'total' form. Relative metal availability, was greater for Zn, Cu, Pb and Cd than Ni in the greenhouse soil. Relative metal availability order of metals in MSW treated soils were Zn>Pb>Cu>Cd>Ni. Relative availability of metals was the least for control treatment, and was the highest for MSW₂₀₀ treatment.

Although total concentrations of all metals were found below the pollutant limits, it can be seen that the increase in DTPA-extractable fractions was more marked than those of total concentrations. These results support the hypothesis [15] that metals added with sewage sludge or other organic wastes may be more mobile in soil than native metals.

IV. CONCLUSION

The results of the present study indicated that soil application of MSW compost increased total and DTPA extractable levels of Zn, Cu, Pb, Ni and Cd in the soil compared with the control. Although there was no phytotoxicity on the plant growth by MSW compost treatments, due to cumulative accumulation and also high bioavailability of metals, plant metal content excessed safety food metal limits. The findings on the effects of heavy metal accumulation in the soil as a result of successive applications of MSW compost to greenhouse soil could be thought a noticeable evidence for the safety reuse concerns of MSW compost.

In the future studies biosolid cleaning technologies involving phytoremediation technics should be taken into

consideration for the composting and safety usage of MSW compost and other biosolids.

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| Treatments | Zn | | Cu | | | Ni | | Pb | Cd | |
|-----------------------|--------------|-------|--------------|------|--------|-------|--------|--------|-----------|---------|
| Treatments | Leaves Fruit | | Leaves Fruit | | Leaves | Fruit | Leaves | Fruit | Leaves | Fruit |
| Control | 44 c | 4.4 c | 11 c | 7 b | 1.6 c | 0.6 c | 3.1 c | 0.4 c | 0.1 b | <0.02 b |
| MSW_{100} | 62 b | 6.7 b | 16 b | 7 b | 4.4 b | 1.8 b | 14.7 b | 3.1 b | 0.8 a | 0.05 a |
| MSW ₂₀₀ | 82 a | 7.8 a | 17 a | 10 a | 7.2 a | 3.4 a | 26.2 a | 11.3 a | 1.1 a | 0.09 a |
| Significance | ** | ** | ** | ** | * | * | ** | ** | * | * |
| Background level [8] | 40 40 | | 8 8 | | 2 2 | | 3 3 | | <0,5 <0.5 | |
| Phytotoxic level [10] | 100- 400 | | 20-100 | | 10-100 | | 30-300 | 1 | 5-30 | |

TABLE II. HEAVY METAL CONTENT IN THE LEAVES AND FRUITS OF CUCUMBER GROWN IN MSW COMPOST APPLIED SOIL

All data are in mg kg⁻¹ dry matter weight. Means followed by the same letter are not statistically different (P ≤ 0.05), *: Significant with P ≤ 0.05 ; **: Significant with $P \le 0.01$

TABLE III. TOTAL AND EDTA EXTRACTABLE CONCENTRATIONS OF HEAVY METALS IN THE GREENHOUSE SOIL SAMPLED IN 2009, AFTER HARVEST OF CUCUMBER.

| Treatments | Zn | | | Cu | | | Ni | | | Pb | | | Cd | | |
|--------------|-------|------|------|-------|------|------|-------|--------|-----|-------|------|------|--------|--------|-----|
| Treatments | Total | DTPA | D/T | Total | DTPA | D/T | Total | DTPA | D/T | Total | DTPA | D/T | Total | DTPA | D/T |
| Control | 77 c | 15 c | 19 c | 19 b | 4 b | 21 b | 17 b | 0.69 b | 4 | 39 c | 7 c | 18 c | <0.02 | <0.02 | - |
| MSW_{100} | 114 b | 25 b | 22 b | 27 a | 7 a | 26 a | 22 a | 0.87 a | 4 | 49 b | 16 b | 33 b | 0.09 b | 0.02 b | 22 |
| MSW200 | 161 a | 77 a | 48 a | 28 a | 8 a | 28 a | 23 a | 0.92 a | 4 | 59 a | 22 a | 37 a | 016 a | 0.07 a | 44 |
| Significance | ** | ** | ** | * | ** | * | * | * | NS | ** | ** | ** | * | * | NS |

All data are in mg kg⁻¹ as dry weight basis. Means followed by the same letter are not statistically different (P \leq 0.05), *: Significant with P \leq 0.05; **: Significant with P \leq 0.01; NS: not significant, D/T: DTPA/Total concentration rate (%)