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Heavy metal input to agricultural soils from irrigation with treated wastewater: Insight from Pb isotopes

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A major objective of the EU FP6 project SAFIR was to overcome certain drawbacks of wastewater reuse through the development of a new irrigation technology combining small-scale modular water treatment plants on farm level and improved irrigation hardware, in the aim to lower the risks related to low quality water and to increase water use efficiency. This innovative technology was tested in several hydro-climatic contexts (Crete, Italy, Serbia, China) on experimental irrigated tomato and potato fields. Here we present the heavy metal variations in soil after medium-term (3 irrigation seasons from 2006-2008) use of treated municipal wastewater with a special focus on lead and lead isotope signatures. The experimental site is located in Chania, Crete. A matrix of plots were irrigated, combining different water qualities (secondary, primary treated wastewater, tap water, partially spiked with heavy metals, going through newly developed tertiary treatment systems) with different irrigation strategies (surface and subsurface drip irrigation combined with full irrigation and partial root drying).

In order to assess small scale heavy metal distribution around a drip emitter, Pb isotope tracing was used, combined with selective extraction. The sampling for Pb isotope fingerprinting was performed after the 3^{rd} season of ww-irrigation on a lateral profile from a drip irrigator (half distance between drip lines, i.e. 50cm) and three depth intervals (0-10, 10-20, 20-40 cm). These samples were lixiviated through a 3 step selective extraction procedure giving rise to the bio-accessible, mobile and residual fraction: $CaCl_2$/$NaNO_3$ (bio-accessible fraction), DPTA (mobile fraction), total acid attack (residual fraction). Those samples were analysed for trace elements (including heavy metals) and major inorganic compounds by ICP-MS. The extracted fractions were then analysed by Thermal Ionisation Mass Spectrometry (TIMS) for their lead isotope fingerprints (^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb). These fingerprints allowed discriminating geogenic (rock-derived) and anthropogenic lead fractions. Both the bulk initial soil before any wastewater irrigation (sampled in 2006) and the residual fractions of all analysed samples have a typical signature of uncontaminated background soils (Erel *et al*., 1997 and references therein). Mobile fractions fall on a mixing line with an endmember comparable to Israeli aerosols, themselves dominated by gasoline lead (Erel *et al*., 2006). Due to labour, this signature can be found throughout the root zone. This shift from background values can be explained by the proximity (<500 m) of a motorway to the experimental plots. The wastewater, spiked with synthetic PbCl2, has a signature clearly distinct from the mixing line, both in the ^{208}Pb/^{206}Pb *vs.* ^{206}Pb/^{207}Pb diagram and in the ^{207}Pb/^{204}Pb *vs.* ^{206}Pb/^{204}Pb diagram, falling in the field of Peri-Mediterranean Pb-ores (Luck & Ben Othman, 2002). Any significant irrigation water input to the mobile fraction could be excluded on the basis of the isotope fingerprint of the $PbCl_2$ used for spiking the irrigation water. Nevertheless, the lead isotopic composition in the bio-accessible fraction ($CaCl_2$/$NaNO_3$ extraction) could not be determined due to very low Pb concentrations in the leachate.

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