

Authors comments to Reviewer # 2:

**Isotropic 2-D soil can be fully described by a randomly selected 1-D variogram. The following has been added to the *revised MS*:**

We here introduce the variographic approach mainly for the cases of 1-D and 2-D as a means of characterising the heterogeneity in the X-Y plane. Compared to the typical major variability in the Z-direction of soil depth profiles (soil horizons, layers, geological formations), the linear (1-D) or 2-D heterogeneity *within* soil horizons is significantly smaller, although this is exactly the kind of heterogeneity the present study aims at controlling. Contrary to depth profile zonation a.o. the within-horizon 1-D and 2-D heterogeneity complies with the requirements of both TOS and geostatistics, i.e. spatial heterogeneity can be modelled variographically w.r.t. a physically meaningful *average level* (the inherent *stationarity assumption* in geostatistics), e.g. Fig.s 2-5. It is not meaningful to apply variographic characterisation on measurement series which contain discontinuous shifts, upsets or other disrupt, level changes, as is the prime characteristic of soil depth zonations. The geostatistical tradition of modelling 2-D patterns based on projection onto a 1-D transect can also be debated<sup>1</sup>. In the present context all *isotropic* 2-D heterogeneity patterns can be characterised comprehensively by a *randomly selected* 1-D direction (transect). In all sampling operations there should pererentially always be some sort of random selection involved, unless compelling geo-science reasons exists for choosing a direction related to the genesis of the specific heterogeneity met with, e.g. choosing a 1-D transect either along a dominant plow direction.

**Augmented description of laboratory sub-sampling and mass reduction. The following has been added to the *revised MS*:**

After the stored samples were thawed and accommodated for 20 °C for a week, before being processed further. The primary field sample size (200-300 gram) must be reduced to the analytical sample size (1-2 gram), not at all a trivial mass-handling issue. In order to provide representative sub-samples, TOS principles were applied scrupulously to all mass reduction steps. Thus samples were dried and macerated, or ground, where appropriate, and subsequently deployed in a longitudinal tray, forming a 1-D lot, using the soil-adapted bed-blending/cross-cut reclaiming technique described in detail in (Petersen et al. 2004) and Kardanpour et al. (2015b). These pre-blended micro-beds were cut by 10 randomly selected transverse increments along the elongated dimension which were aggregated, resulting in subsamples of 20-30 gram each. The exact same procedure was repeated in a secondary mass reduction step ending up with the final analytical mass (2 gram) for the wet samples analyses. This procedure has been honed to full representativity in the course of this project specifically so as to do away with all of the post-primary-sampling errors in order better to be able to focus in the latter and the variogram deployment, *ibid*.

The remainders of the secondary sub-samples were air-dried for four days in lab temperature (20 °C), to be used in parallel sorption experiments. As a further scale-down iteration, a similar bed-blending/cross-cut reclaiming were used to provide analytical samples of 2 gram, also based on 10 increments each.

Kardanpour et al. (2015b) describe the “from-field-sampling-to-aliquot” pathway in full details, complete with an exhaustive pictorial exposé.

---

<sup>1</sup> The present authors do not wish to reject the 2-D geostatistical tradition with this statement, but in relation to the present matters this issue is better deferred to another occasion in which the 2-D modelling issue can be presented and discussed in full - this issue is a legitimate and interesting area for a fruitful debate. Entering into a 3-D geostatistical modelling realm, there are also here issues that in need of further discussion, e.g. the required minimum number of samples (measurements) needed for meaningful, and stable variogram calculation. The present foray only aims at presenting the power of a simple 1-D variogram characterisation operator based on TOS, upon which several versions of potential follow-up generalisations to 2-D and 3-D cases may be entertained.

Kardanpour, Z, Jakobsen, O.S. & Esbensen, K.H. (2015b) Counteracting soil heterogeneity sampling for environmental studies (pesticide residues, contaminants transformation) - TOS is critical. Proceedings 7.th World Conference on Sampling and Blending (WCSB7), p.205-209.

**Augmented argumentation re. primary sampling a.o. The following has been added:**

The primary sampling was specifically intended to correspond to current sampling traditions in the soil and microbiology communities. In other studies efforts have been made to optimize each individual field sample, for example with respect to the famous “Gy’s formula”, from which control over the so-called Fundamental Sampling Error is often sought. However, in the present study it is a major point to outline how the variographic approach a.o. lead to a procedure with which to characterize the magnitude of the total sampling-plus-analytical error and thus to be warned of the need to control (better) all the inherent sampling errors, see e.g. DS 3077 (2013) for a comprehensive introduction.

**Rational for a central Roman Square etc. The following has been added:**

The experimental design allows comparison of the small-scale and large-scale variability. All profiles can for example be directly compared with the level and variation at the small-scale experiment, by the pertinent mean  $\pm 2$  SD. This is just for visual orientation however and not to be confused with the nugget effect, a much more general characterisation of the small(est) scale variability pertaining to below lag = 1, summing up and averaging this information for all the sample pairs in the transect.

**General rationale of paper; further elucidation. The following has been added:**

In cases where the next step in studies might be assessment of the main factors driving the spatial heterogeneity of soil contamination analytes for example, the 1-D (or 2-D X-Y) approach advocated here, will only serve as a basis for proper selection of experimental material to be taken to the laboratory - upon which further considerations will focus on, say, the potential factors involved in contaminant input and transport a.o. Note that these latter processes manifest themselves primarily in the Z-direction, where it is by no means a given that application of the same variographic approach (or geostatistical modelling) will necessary give meaningful results.

**Reviewer #2 did not like the use of “profile” in the meaning of a horizontal 1-D profile.**

We agree that this is confusing in the soil science world. We have replaced all such use with the “transect”.