Environmental Science & Technology

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African Soils Are Also under Threat: Occurrence of Pesticides in Agricultural Soils and Implications

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Cite This: *Environ. Sci. Technol.* 2026, 60, 17446−17460

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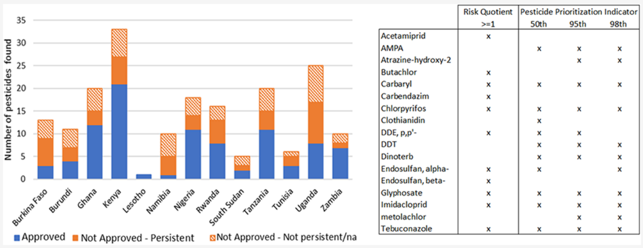

ABSTRACT: Pollution is recognized as a major threat to soil health; however, significant gaps persist regarding the distribution and risks of chemicals in developing regions. This study investigated the occurrence of pesticide residues in African agricultural soils. Between 2022 and 2024, we collected 272 topsoil samples from 13 countries representing diverse farming systems and analyzed them for 153 pesticide residues, including active ingredients and their metabolites. Pesticides were detected in 216 samples, with 63 different residues identified, several of which were unregistered in the countries where they were found and some banned in the European Union. The most frequently detected compounds were dinoterb, DDE-p,p′, chlorpyrifos, metolachlor, atrazine-hydroxy-2, AMPA, and imidacloprid, all of which are substances of environmental, ecotoxicological, or toxicological concern. Individual pesticide concentrations ranged from <0.001 to 4.896 mg kg^{-1}, and the total pesticide load reached 6.998 mg kg^{-1}. At least 25% of the sampling locations are expected to present a high ecological risk (RQ ≥ 1), although data gaps for some substances limit assessment accuracy. Eleven pesticides exceeded the risk thresholds in at least one sample, with chlorpyrifos driving the most exceedances. A monitoring- and hazard-based prioritization approach highlighted additional high-concern compounds, notably dinoterb and glyphosate/AMPA. Overall, the findings reveal the significant environmental impacts of agricultural intensification and highlight regulatory challenges related to pesticide use across Africa.

KEYWORDS: *soil health indicators, monitoring, risk assessment, prioritization, agricultural intensification*

1. INTRODUCTION

According to Tang et al.,[1] 64% of global agricultural land is at risk of pesticide pollution by more than one active ingredient, and 31% is at a high risk. Nearly 20% of high-risk areas are in low- and lower-middle-income nations. The validation of these and other modeling or machine learning-based estimates, along with their underlying assumptions, such as pesticide levels in soil, using measurements of field samples, remains particularly challenging. This is mostly because of the limited monitoring of pesticide levels in the environment after approval for use in the country, the high costs, and the analytical challenges related to such a diverse group of contaminants in such a complex matrix. The extent of soil contamination in Africa is largely unknown,[2] but it is likely to be of concern due to a lack of pesticide management and regulations in many countries and poor law enforcement about the use of hazardous pesticides.[3] Existing studies monitoring pesticides in soils cover a limited number of pesticides or a relatively small area. These studies vary greatly in sampling time, sampling strategy, and analytical methods, hampering an overview of the occurrence and levels of residues in agricultural soils.[4] At the same time, available studies stress the omnipresence of mixtures of pesticide residues and alert to the dangers of pesticides to soil health[5] and connected compartments.[6,7] Evidence of the negative effects of pesticides on nontarget organisms, global biodiversity, and the environment is abundant, covering multiple groups of organisms, ecosystems, and climatic conditions.[8,9] At the soil level, pesticide use is often associated with lower soil biodiversity, a condition exacerbated by the use of broad-spectrum substances and the application of multiple substances.[10,11] Several pesticide residues are persistent in soil and toxic to soil organisms even at recommended application rates.[12] Adverse effects may also result from the accumulation of pesticide residues in organisms or via pesticide-driven alterations in habitat or ecosystem structure.[11,13] Unraveling and monitoring pesticide contamination of African soils is, therefore, urgently needed to assess and monitor their quality and health. This information will support policymakers in pesticide-related actions.[14]

Received: October 31, 2025
Revised: April 22, 2026
Accepted: April 22, 2026
Published: June 8, 2026

Monitoring data in soils, as contextualized above, is rather fragmented and especially scarce in Africa.[4] A relatively high number of African studies were identified in the literature (n = 88), covering 25 African countries. However, a closer look shows that (i) most of the data originates from 3 countries (Nigeria, Tanzania, Kenya), (ii) most studies focus on a few and often the same compounds (mostly organochlorinated pesticides), (iii) data on more modern pesticides such as neonicotinoids (e.g., imidacloprid, acetamiprid, thiamethoxam) are particularly scarce, and (iv) the data are rather old (most are prior to 2015). In line with these, the overall aim of this study was to provide a harmonized, updated, and comprehensive overview of soil contamination by pesticide residues in Africa and explore the possible implications of such findings to soil health via hazard-based indicators. These indicators were also used to identify the compounds of the highest interest for further investigation and potentially priority targets for use and risk reduction actions.

2. MATERIALS AND METHODS

2.1. Soil Samples

This study was embedded in the European Union Horizon 2020-funded Soils4Africa project. The Soils4Africa project is a pan-African research initiative aimed at improving access to harmonized and up-to-date soil information to support sustainable land management, climate resilience, and agricultural productivity across Africa. To achieve these objectives, the project conducted a continent-wide field survey to generate new soil data, providing baselines of the state of soil of Africa's agricultural soils for future monitoring, and established a continental soil information system to disseminate these data. Pesticide analyses were planned for topsoil samples taken from 300 sites, covering a mixture of real-field conditions: crops, farming systems, and pesticide use regimes. In the end, we were able to collect and analyze samples from 272 locations, a sample size that is in line with that of Silva et al.,[15] with the same exploratory assessment character but for Europe. Each of the 272 samples was a composite, unit-volume sample and represented the upper 5 cm of the soil surface, where pesticides tend to accumulate.[16,17] Field surveyors were instructed to, at each pesticide sampling location, combine the contents of 3 metallic rings with a volume of 100 cm^3 into a plastic bag, which was thereafter considered the pesticide sample. Surveyors removed stones and other gross materials from the samples, kept the samples in cool and dark conditions, and shipped them as soon as possible to the collection point for soil samples of the Soils4Africa project in South Africa. The pesticide samples were deep-frozen (−20 °C) at the collection point and shipped to Wageningen University and Research, in The Netherlands, as soon as a reasonable number of samples were available for shipment.

The 272 pesticide samples originated from 13 countries: Burkina Faso (BF), Burundi (BI), Ghana (GH), Kenya (KE), Lesotho (LS), Namibia (NA), Nigeria (NG), Rwanda (RW), South Sudan (SS), Tanzania (TZ), Tunisia (TN), Uganda (UG), and Zambia (ZM). The number of samples per country ranged from 2 to 47. The two-letter country code combined with the order in which the samples were received at the African collection point was used to derive pseudoanonymized sample identifiers. Pesticide samples were collected over a 2-year span from October 2022 to September 2024. Almost all pesticide sampling locations showed signs of soil erosion and grazing. The water supply in these locations is primarily rainfed. Over 60% of the sampling locations showed signs of past or recent tillage. Tillage was often done manually. Out of the 272 samples, 188 originated from cultivated and managed land (terrestrial area), 12 from cultivated aquatic or regularly flooded areas, and 72 from seminatural vegetation (which has been used for agriculture in the past but is currently not cultivated). Cereals were the most represented crop (n = 105), followed by dry pulses (n = 25), roots and tubers (n = 20), and fruits (n = 15; Table S1).

2.2. Pesticide Determinations

Pesticide determinations were done using standardized and traceable methods similar to those described by Silva et al.[15] The list of analytes was defined based on evidence and the likelihood of occurrence of pesticide residues in African soils (Figure 1). The occurrence of evidence was assessed by a literature review and full screening of about 10% of the samples. Full screen was used as a quality control measure to ensure that no compounds were overlooked and to compensate for possible limitations and bias of the published results. The "likely to occur" residues were obtained from databases on pesticide use and from a simple pesticide questionnaire used in the field on past and current pesticide use. Analytical capacity was explored next, with a preference to multiresidue methods involving a generic extraction and cleanup approach. The QuEChERS (Quick, Easy, Cheap, Effective, Rugged, Safe) method was adapted for soil samples, using the steps of Silva et al.[15] QuEChERS was followed by using liquid and gas chromatography analyses combined with mass spectrometric detection: LC-MS/MS is liquid chromatography-tandem mass spectrometry, and GC-MS/MS is gas chromatography-tandem mass spectrometry. Considering the global relevance and interest in glyphosate, an additional single-residue method was used for the analysis of glyphosate and its main metabolite aminomethylphosphonic acid (AMPA). The nature of the pesticide, analytical performance, and availability of standards were considered. A final list of 153 pesticides was compiled and analyzed in all soil samples. This list included 132 active ingredients, 20 metabolites, and 1 synergist (Figure 1).

Quality control of the analytical work was performed according to the SANTE 11312/2021 guidance document on analytical quality control and method validation procedures for pesticide residue analysis in food and feed.[18] Quality control measures involved the use of multiple pesticide calibration standards of reference and isotope-labeled internal standards, and fortified soils. This was done to assess the linearity of the response versus the concentration in the calibration curves, recoveries, and secure multistep compound identification. More details on the Quality Assurance/Quality Control used in the lab are available in Silva et al.[15] Limits of quantification (LOQ) were used as the reporting limits. LOQs ranged from 1 to 10 $\mu g\ kg^{-1}$ for pesticide residues, as follows: 1 $\mu g\ kg^{-1}$ for 131 residues, 2.5 $\mu g\ kg^{-1}$ for 6 residues, 5 $\mu g\ kg^{-1}$ for 11 residues, and 10 $\mu g\ kg^{-1}$ for 5 residues. See Table S2 (Wageningen University & Research (2025). Soils4Africa - Pesticide dataset (Version v01) [Data set]. Zenodo. https://doi.org/10.5281/zenodo.177779527) for the LOQ compound-specific information. Pesticide content is always expressed as soil dry matter.

2.3. Contextual Data

As mentioned above, a simple questionnaire was prepared to retrieve information about current and past pesticide use at the pesticide sample locations. Additional information about the samples and sampling locations was provided by the Soils4Africa project. Sample indicators included soil pH, soil organic carbon, and clay, silt, and sand content. Sampling location indicators included slope, existence of plowing, erosion, and grazing signs, tillage mode, and water supply method. The basic properties of the 153 analytes were compiled (Table S3) to support compound-specific evaluations. The data was extracted from the Pesticide Properties DataBase (PPDB)[19] or Bio-Pesticides DataBase;[20] see Lewis et al.[21] for details on these databases. The data retrieved included (i) solubility in water at 20 °C; (ii) octanol−water partition coefficient at pH 7, 20 °C, and log P; (iii) soil degradation rates; (iv) soil adsorption coefficient, K_d (distribution coefficient), and K_{oc} (coefficient normalized to the organic carbon content), (v) vapor pressure at 20 °C, and (vi) Henry's law constant at 25 °C. The environmental, ecotoxicological, and toxicological profiles of the most common pesticide residues in soil were retrieved from PPDB. The pesticides found in the soil samples were aggregated at the country level and compared to the list of pesticides registered in the respective country (see specific links to the list of registered products in Supporting Information, under Tables S9−S21). The approval status in the EU was also collected from the EU Pesticide

https://doi.org/10.1021/acs.est.5c15564
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Table 1. Overview of Pesticide-Specific Findings: Frequency and Levels, and Detection vs National Approval Status[a]

	Compound	found in N samples	found in N countries	mean, of positive	max. content	BF (N=10)	BI (N=10)	GH (N=18)	KE (N=47)	LS (N=2)	NA (N=10)	NG (N=30)	RW (N=10)	SS (N=17)	TZ (N=33)	TN (N=18)	UG (N=47)	ZM (N=20)	EU Status?
	Number PESTICIDES DETECTED/country					13	11	20	33	1	10	18	16	5	20	6	25	10	
	Number NOT APPROVED detected/country					10	7	8	12	0	9	7	8	3	9	3	17	3	
	Number NOT APPROVED detected/country/PERSISTENT					6	3	3	6	0	4	3	5	1	4	2	9	1	
1	Dinoterb	76	11	0.020	0.470	F	F	F	F		F	F		F	F	F	F	F	NOT Ap.
2	DDE p,p'	58	8	0.007	0.143	F	F		F				F		F	F	F	F	-
3	Chlorpyrifos	55	7	0.004	0.014		F	F	F			F		F			F	F	NOT Ap.
4	Metolachlor (S)	54	8	0.003	0.036			F	F	F	F	F			F		F	F	Approved
5	Atrazine-hydroxy-2	38	8	0.006	0.030	F		F	F		F	F		F	F		F		-
6	AMPA	37	6	0.171	2.093	F		F	F			F			F		F		-
7	Imidacloprid	32	6	0.006	0.048			F	F				F		F		F	F	NOT Ap.
8	Carbaryl	24	3	0.031	0.103						F						F	F	NOT Ap.
9	Clothianidin	22	4	0.003	0.023				F				F		F		F		NOT Ap.
10	Pendimethalin	21	5	0.004	0.065			F	F		F	F			F				Approved
11	Endosulfan, alpha-	20	5	0.007	0.038	F		F				F	F		F				NOT Ap.
12	Pentachloroanisole	20	4	0.003	0.009		F	F	F								F		-
13	Endrin	17	4	0.005	0.046				F				F			F	F		NOT Ap.
14	Pentachloroaniline	17	4	0.002	0.004		F							F	F		F		-
15	Endosulfan, beta-	16	5	0.004	0.022	F	F		F						F		F		NOT Ap.
16	Tebuconazole	16	4	0.292	1.019				F			F	F			F			Approved
17	Permethrin	15	8	0.012	0.053	F	F	F			F		F		F	F	F		NOT Ap.
18	DDT p,p'	14	3	0.035	0.104		F		F								F		NOT Ap.
19	Dieldrin	14	2	0.001	0.003	F											F		NOT Ap.
20	Metalaxyl	14	3	0.004	0.021							F			F			F	Approved
21	Glyphosate	13	4	0.465	4.896			F	F			F					F		Approved
22	Transfluthrin	11	2	0.003	0.005	F						F							NOT Ap.
23	Pirimiphos-methyl	10	4	0.003	0.009								F	F			F	F	Approved
24	Carbendazim	9	4	0.006	0.026				F				F		F		F		NOT Ap.
25	Bifenthrin	7	2	0.002	0.002			F					F						NOT Ap.
26	Malathion	7	4	0.005	0.013				F				F		F			F	Approved
27	Propoxur	7	3	0.001	0.002	F		F							F				NOT Ap.
28	Acetamiprid	6	2	0.004	0.012			F	F										Approved
29	Atrazine	6	3	0.002	0.004			F	F			F							NOT Ap.
30	Azoxystrobin	6	2	0.009	0.015				F			F							Approved
31	Thiamethoxam	6	3	0.004	0.013				F						F		F		NOT Ap.
32	Imidacloprid desnitro	5	2	0.004	0.006			F	F										-
33	Acetochlor	4	3	0.002	0.003				F								F	F	NOT Ap.
34	DDD p,p'	4	3	0.003	0.004	F							F				F		-
35	DDE, o,p'	4	3	0.003	0.007				F				F				F		-
36	Diazinon	4	1	0.004	0.010												F		NOT Ap.
37	Difenoconazole	4	2	0.007	0.012				F							F			Approved
38	HCH, alpha-	4	3	0.005	0.012	F			F				F						NOT Ap.
39	Hexachlorobenzene	4	1	0.022	0.039			F											NOT Ap.
40	Chlorfenapyr	3	2	0.003	0.004							F			F				NOT Ap.
41	Flubendiamide	3	1	0.004	0.010				F										Approved
42	Fluopyram	3	2	0.009	0.024				F		F								Approved
43	Allethrin	2	2	0.008	0.010			F				F							NOT Ap.
44	Butachlor	2	2	0.004	0.007	F			F										NOT Ap.
45	Diuron	2	2	0.003	0.004							F					F		NOT Ap.
46	Terbuthylazine	2	1	0.002	0.003				F										Approved
47	2,4-D (free)	1	1	0.003	0.003			F											Approved
48	Alachlor	1	1	0.001	0.001							F							NOT Ap.
49	Boscalid	1	1	0.008	0.008						F								Approved
50	Carbofuran, Keto	1	1	0.002	0.002		F												-
51	Cyproconazole	1	1	0.008	0.008				F										NOT Ap.
52	Denatonium benzoate	1	1	0.010	0.010						F								NOT Ap.
53	Endosulfan ether	1	1	0.004	0.004								F						NOT Ap.
54	Fenpropathrin	1	1	0.001	0.001										F				NOT Ap.
55	HCH delta-	1	1	0.002	0.002		F												NOT Ap.
56	Lambda-Cyhalothrin	1	1	0.004	0.004			F											Approved
57	Metribuzin	1	1	0.002	0.002				F										Approved
58	Myclobutanil	1	1	0.002	0.002							F							NOT Ap.
59	Pentachlorobenzene	1	1	0.001	0.001										F				-
60	Piperonyl butoxide	1	1	0.004	0.004								F						
61	Profenofos	1	1	0.001	0.001		F												NOT Ap.
62	Propiconazole	1	1	0.002	0.002				F										NOT Ap.
63	Spiroxamine	1	1	0.001	0.001						F								Approved

[a]The table covers the 63 pesticides found in the soil, sorted from the most to the least common compounds. Color gradients were added to the frequency and level columns for quick comparisons among the pesticides. Dark blue = active ingredients approved in the country, light blue = active ingredients not approved in the country, white = not applicable/metabolite. Concentrations were rounded to three decimal places. F = compound found in a certain country, BF = Burkina Faso, BI = Burundi, GH = Ghana, KE = Kenya, LS = Lesotho, NA = Namibia, NG = Nigeria, RW = Rwanda, SS = South Sudan, TZ = Tanzania, TN = Tunisia, UG = Uganda, and ZM = Zambia.

database.[22] EU Pesticide approval status refers to 1 January 2022, right before the sampling campaign.

2.4. Risk Quotients and Pesticide Prioritization Indicators

Terrestrial ecotoxicological data were compiled for pesticides found in the soil (Table S4). Three databases were consulted for such a

https://doi.org/10.1021/acs.est.5c15564
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compilation: PPDB, OpenFoodTox,[23] and the ECOTOX database.[24] In the PPDB, an R script was used to extract LC_{50} (the Lethal Concentration for 50% of the population) and NOEC (No Observed Effect Concentration) values for reproduction in earthworms and collembolans. Similar data were manually retrieved in the other two databases, using CAS numbers as pesticide identifiers. In OpenFoodTox, the following soil invertebrate species were selected: *Eisenia fetida, Lumbricus terrestris, Aporrectodea caliginosa, Folsomia candida*, and *Hypoaspis aculeifer*. In the ECOTOX database, a larger species coverage was possible: *E. fetida, E. andrei, A. caliginosa, A. longa, Perionyx excavatus, Allolobophora icterica, L. rubellus, L. terrestris, Enchytraeus albidus, F. candida, F. fimetaria, Heteromurus nitidus*, and *H. aculeifer*. The data retrieved from the three databases were merged, and duplicates were removed. Soil toxicity data were obtained for 49 out of the 63 detected pesticides. Several steps were taken to fill these gaps. First, for 5 metabolites (atrazine-hydroxy-2; carbofuran-keto; endosulfan ether, imidacloprid desnitro, and DDD-p,p'), the lowest toxicity value of the corresponding parent compound was used. For HCH (hexachlorocyclohexane)-α and HCH-delta, the data of the isomer HCH-γ was used, and for DDE-p,p', the data of DDE-o,p' was used. European Food Safety Authority (EFSA) publications were reviewed to fill the remaining data gaps, but additional information was found only for pirimiphos-methyl. No data were found for chlorfenapyr, denatonium, dinoterb, pentachloroanisole, and piperonyl butoxide. Extrapolation from aquatic ecotoxicological data was explored but not further pursued because of the limited or complete lack of data.

The compiled ecotoxicological information was used in the calculation of the Risk Quotients (RQ) and Pesticide Prioritization Indicator (PPI). The RQ, a common approach used in monitoring studies, informs site-specific risks and their main drivers. RQ was calculated for each analytical finding (≥LOQ) as the ratio of the measured concentration in soil to the corresponding Predicted No Effect Concentrations (PNECs). PNECS were obtained for each pesticide as the ratio of its lowest ecotoxicity value to the respective assessment factor (AF), following the approaches of Vašíčková et al.[25] and Pelosi et al.[26] If only acute LC_{50} data were available, an AF of 1000 was applied; if both LC_{50} and NOEC values were available, an AF of 10 was used. For pesticides with high RQ and ecotoxicity data retrieved from the ECOTOX database, the CRED (Criteria for reporting and evaluating ecotoxicity data) evaluation was applied.[27] Nine studies were assessed; those failing CRED were excluded, and the next lowest toxicity value was selected. This process was repeated until a valid study was found or the lowest values were from PPDB or OpenFoodTox. For each soil sample, the total risk quotient ($\sum RQ$) was determined as the sum of the RQ for all pesticides found in the sample. The total risk quotient was then classified as negligible risk ($\Sigma RQ < 0.01$), low risk ($0.01 \leq \Sigma RQ < 0.1$), medium risk ($0.1 \leq \Sigma RQ < 1$), and high risk ($\Sigma RQ \geq 1$).[25] The PPI, a pesticide-specific, occurrence and hazard-based indicator, was applied, as originally proposed by Silva et al.,[28] to identify the compounds of highest interest for pesticide use and risk reduction actions. The PPI formula accounts for the frequency of detection of the pesticide across soil samples, the median level of the pesticide in soil, and the highest hazard score of the pesticide among soil organisms and ecotoxicological endpoints. A 0/1/2/3 hazard score is attributed to low/no data/moderate/high toxicity classes, respectively. The values in Table S4 were attributed to these toxicity classes, in line with the PPDB toxicity thresholds.

2.5. Further Data Analyses

Descriptive statistics were performed at the continent, country, crop, and pesticide levels. These include analyses of the number of residues in the samples, the residues found (and not found) in the soil, the frequency and levels in the soil, and the pesticide profile per sample. As mentioned above, LOQs were used as reporting limits, which means that only pesticide contents equal to or above the pesticide LOQs were considered in the metrics, including the statistical analyses. Entries where the pesticide level was below the LOQ were left empty. The sample profile was defined as a set of pesticides ≥ LOQ. Sample profiles were compared using their full composition signatures and were checked for partial compositional concordance.

Sample findings were represented spatially to highlight the coverage of the data set and explore the occurrence of hotspots and spatial clusters. In other figures, to simplify comparisons, the number of pesticide residues in the soil and the total pesticide content in the soil were aggregated by classes: "0, 1, 2−5, 6−10, >10 residues" and "No residues ≥ LOQ, ≥LOQ−0.05, ≥0.05−0.15, ≥0.15−0.5, ≥0.5−1, ≥1 mg kg^{-1}", respectively. These classes are commonly used in European studies.[15,29] Nonparametric Mann−Whitney tests were used to compare the total pesticide content in soil among different countries. This was done for the complete data set and the cereal class, which had the highest number of samples. A significance level of 0.05 was used in the tests. Spearman's rank correlations were used to explore the possible relationships between the content of pesticides in the soil and basic soil properties.

3. RESULTS

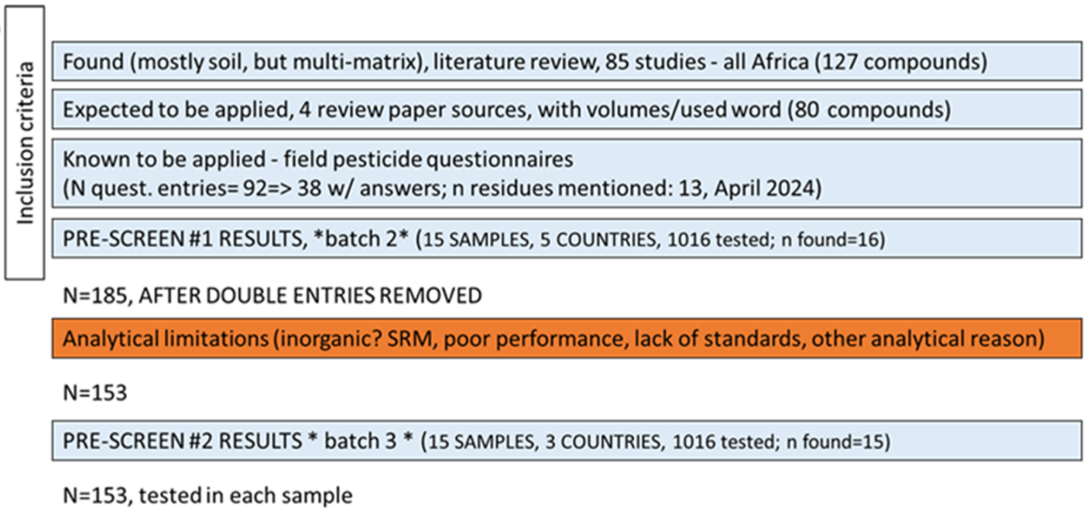
3.1. Overview of Pesticide Findings

3.1.1. Contaminated/"Clean" Samples. Out of the 272 samples (21%) tested, 56 samples were "pesticide-free", 53 samples (19%) had one pesticide residue, and 163 samples (60%) had mixtures of pesticide residues (Figure 2). Simple mixtures of 2 to 5 residues were the rule. A few samples had more than 10 pesticide residues, with a maximum of 14 pesticide residues per sample: RW_06 (11 residues), UG_44 (11 residues), TZ_18 (12 residues), UG_45 (12 residues), UG_46 (12 residues), KE_01 (14 residues), and UG_41 (14 residues). A total of 153 pesticide profiles were detected among the soil samples, of which 139 were found in a single soil sample. The most common profile in our data set was a two-compound "chlorpyrifos + DDE-p,p" mixture, found in 6 samples. The "metalaxyl + metolachlor + carbaryl + chlorpyrifos" and "metolachlor + carbaryl" mixtures were found in 3 samples. Eleven mixtures were detected in two samples: "metolachlor + pendimethalin + atrazine-hydroxy-2 + dinoterb", "imidacloprid + chlorpyrifos + DDE-p,p", "metalaxyl + metolachlor + carbaryl + dinoterb", "clothianidin + imidacloprid", "metolachlor + carbaryl + dinoterb + chlorpyrifos", "dinoterb + DDE-p,p", "metolachlor + dinoterb", "dinoterb + endosulfan, α-", "dinoterb + transfluthrin", and "dinoterb + pentachloroaniline". As highlighted in this short list, there were common pesticide components among soil samples; see, for instance, the often co-occurrence of endosulfan-α and pentachloroanisole, and of endrin-endosulfan-β, DDT-p,p', and pirimiphos-methyl (Figures S1 and S2).

3.1.2. "Present"/"Absent" Pesticide Residues. Sixty-three pesticide residues were found in the soil samples. Of these, 23 were found in at least ten samples, and the remaining 40 residues were found only occasionally (Table 1). The 63 "found pesticides" list includes 27 insecticides (IN), 12 herbicides (HB), 12 fungicides (FU), 10 metabolites, 1 synergist (piperonyl butoxide), and 1 semiochemical, repellent pesticide (denatonium benzoate). Eight pesticides are PFAS (per- and polyfluoroalkyl substances): bifenthrin, chlorfenapyr, flubendiamide, fluopyram, indoxacarb, lambda-cyhalothrin, thiazopyr, and trifloxystrobin. Thirty-four of the active ingredients found in African soil are not approved for use in Europe: dinoterb (HB), atrazine (HB), acetochlor (HB), butachlor (HB), diuron (HB), alachlor (HB), carbendazim (FU), hexachlorobenzene (FU), cyproconazole (FU), myclobutanil (FU), propiconazole (FU), chlorpyrifos (IN), imidacloprid (IN), carbaryl (IN), clothianidin (IN), endo-

https://doi.org/10.1021/acs.est.5c15564
Environ. Sci. Technol. 2026, 60, 17446−17460

(a)



(b)

LC-MS/MS ESI pos	Abamectin	Chlorotoluron	Fenamiphos	Methamidophos	Pendimethalin	Spirotetramat
	Acetamiprid	Chlorsulfuron	Fenamiphos-sulfoxide	Methidathion	Phenthoate	Spiroxamine
	Aldicarb	Clothianidin	Fenamiphos-sulfone	Methiocarb	Phorate	Tebuconazole
	Atrazine	Cyantraniliprole	Fenthion	Methomyl	Phosalone	Terbufos
	Atrazine, desethyl	Cyproconazole	Flubendiamide	Metolachlor (S)	Piperonyl butoxide	Terbuthylazine
	Atrazine-hydroxy-2	Demeton-s-methyl	Fluopyram	Metrafenone	Pirimicarb	Thiabendazole
	Azoxystrobin	Denathonium benzoat	Formetanate	Metribuzin	Pirimiphos-methyl	Thiacloprid
	Boscalid	Diazinon	Imidacloprid	Mevinphos	Prochloraz	Thiamethoxam
	Buprofezin	Difenoconazole	Imidacloprid, desnitro-	Monocrotophos	Profenofos	Thiazopyr
	Cadusafos	Dimethachlor	Indoxacarb	Monuron	Propargite	Thiodicarb
	Carbaryl	Dimethoate	Isoproturon	Myclobutanil	Propiconazole	Thiophanate-methy
	Carbendazim	Dimethomorph	Linuron	Oryzalin	Propoxur	Triadimefon
	Carbofuran	Disulfoton	Malathion	Oxadixyl	Pyraclostrobin	Trifloxystrobin
	Carbofuran, -3-hydroxy	Diuron	Metalaxyl	Oxamyl	Pyridaphenthion	
	Carbofuran, -keto	Emamectin	Metamitron	Paclobutrazol	Pyrimethanil	
	Cartap	EPTC	Metazachlor	Penconazole	Simazine	
LC-MS/MS ESI neg	2,4-D (free)	Bromoxynil	Dinoterb	MCPA		
GC-MS/MS	Acetochlor	Chlorpyrifos	Dichlorvos	HCH alpha-	Nonachlor (trans)	
	Alachlor	Chlorpyrifos-methyl	Dieldrin	HCH beta-	Parathion	
	Aldrin	Chlorthal-dimethyl	Endosulfan ether	HCH delta-	Parathion-methyl	
	Allethrin	Cypermethrin	Endosulfan alpha-	Lindane	Pentachlorbenzene (PeCB)	
	Azinphos methyl	DDD o,p'	Endosulfan beta-	Heptachlor	Pentachloroaniline	
	Bifenthrin	DDD p,p'	Endosulfan sulphate	Heptachlor endo epoxide (iso A)	Pentachloroanisole	
	Butachlor	DDE p,p'	Endrin	Heptachlor epoxide (iso B)	Permethrin	
	Chlordane cis- (alpha)	DDE, o,p'	Endrin ketone	Hexachlorobenzene	Propanil	
	Chlordane trans- (gamma)	DDT o,p'	Fenitrothion	Hexazinone	Tetrachlorovinphos	
	Chlorfenapyr	DDT p,p'	Fenpropathrin	Lambda-Cyhalothrin	Transfluthrin	
	Chlorpropham	Deltamethrin	Fenvalerate	Mirex		
SRM	Glyphosate	AMPA				

Figure 1. Pesticide-targeted list: (a) steps considered in the preparation of the target list, (b) 153 pesticide residues analyzed in the soil, clustered by analytical method. SRM = single-residue method; AMPA = aminomethylphosphonic acid; DDD = dichlorodiphenyldichloroethane; DDT = dichlorodiphenyltrichloroethane; EPTC = ethyl dipropylthiocarbamate; HCH = hexachlorocyclohexane.

sulfan α- (IN), endrin (IN), endosulfan β (IN), permethrin (IN), DDT-p,p' (IN), dieldrin (IN), transfluthrin (IN), bifenthrin (IN), propoxur (IN), thiamethoxam (IN), diazinon (IN), HCHα (IN), chlorfenapyr (IN), allethrin (IN), endosulfan ether (IN), fenpropathrin (IN), HCH-delta (IN), profenofos (IN), and denatonium benzoate (repellent). Several of the substances found in the soil are not registered/approved for use in the countries where they are found (Table 1). The percentage of nonregistered compounds ranged from 0% in Lesotho to 90% in Namibia. In seven of the 13 countries covered in this study, the percentage of nonregistered pesticides was 50% or more: Burkina Faso, Burundi, Namibia, Rwanda, South Sudan, Tunisia, and Uganda. Nonregistered compounds often had high frequencies and high levels in the soil (see Tables S9–S21). Persistent pesticides comprised 49% of the "detected and "not approved" cases. At the national level, this persistent pesticide contribution ranged between 33% (in South Sudan and Tanzania) and 67% (Tunisia).

The most common residues in African soils were dinoterb (frequency of detection: 28%), DDE-p,p' (21%), chlorpyrifos (20%), metolachlor (20%), atrazine-hydroxy-2 (14%), AMPA (14%), and imidacloprid (12%; Table 1). Most of the compounds in this short list seem to be, or have been, in the past, of rather widespread use. Dinoterb was detected in 11 out of the 13 countries tested and in 9 out of the 12 crop classes. DDE-p,p' and metolachlor were present in 17 out of the 25 "units" assessed (i.e., 13 countries + 12 crops), chlorpyrifos in 16, AMPA in 15, atrazine-hydroxy-2 and imidacloprid in 14 and 13, respectively. The short half-life of dinoterb suggests the recent use of this compound. For the other compounds, with higher half-lifes and high DT_{90} values, interpretations regarding use and legacy effects are more difficult. DDE showed a very high affinity for organic matter, and metolachlor, atrazine-hydroxy-2, AMPA, and imidacloprid were all correlated significantly with the clay content of the soil ($p < 0.05$), which may partially explain the higher than

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Environ. Sci. Technol. 2026, 60, 17446−17460

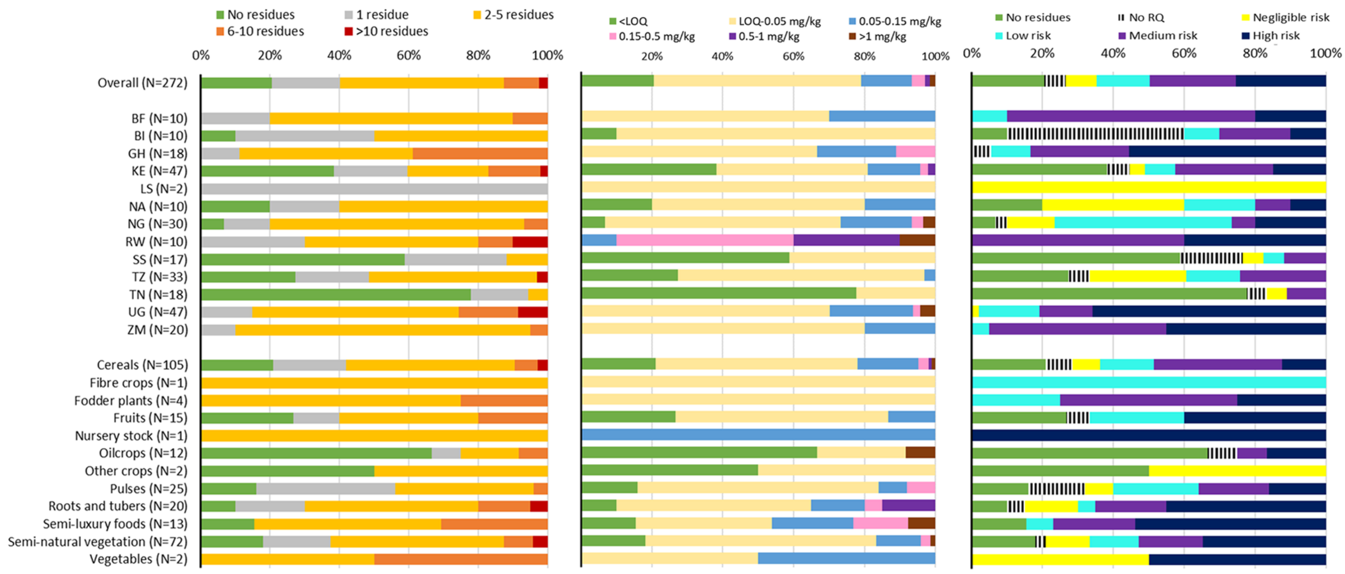

Figure 2. Number of pesticide residues, total pesticide content, and ecological risk in African soil samples. RQ= risk quotient; LOQ= limit of quantification; *N*= number of samples tested. Country abbreviations: Burkina Faso (BF), Burundi (BI), Ghana (GH), Kenya (KE), Lesotho (LS), Namibia (NA), Nigeria (NG), Rwanda (RW), South Sudan (SS), Tanzania (TZ), Tunisia (TN), Uganda (UG), and Zambia (ZM).

expected persistence of some residues (Table S5). Dinoterb, DDE-p,p', and imidacloprid are known to be of high concern based on environmental, ecotoxicological, and toxicological criteria. Atrazine-hydroxy-2 also has a great concern profile but lacks information on environmental impacts and ecotoxicity. Chlorpyrifos, metolachlor, and AMPA are of great concern for some criteria and of medium concern for others (Table S6).

3.1.3. Individual and Total Pesticide Levels. The levels of individual pesticide residues ranged from 0.001 to 4.896 mg kg^{-1}. The mean of the individual pesticide levels ranged from 0.001 to 0.465 mg kg^{-1}, and the median levels ranged from 0.001 to 0.224 mg kg^{-1}. Glyphosate, tebuconazole, and AMPA had the highest content in the soil, with mean values of 0.465, 0.292, and 0.171 mg kg^{-1}, medians of 0.020, 0.224, 0.037, and maximum values of 4.896, 1.019, and 2.093 mg kg^{-1}, respectively. The compounds with the second-highest mean values were DDT-p,p', carbaryl, DDD-p,p', dinoterb, and permethrin. These compounds had mean levels ranging from 0.035 (DDT-p,p') to 0.012 mg kg^{-1} (permethrin). The remaining 55 compounds had mean levels below 0.010 mg kg^{-1}. The compounds with the second-highest maximum values were dinoterb, DDE-p,p', DDT-p,p', and carbaryl (range 0.470−0.103 mg kg^{-1}). Twenty-two compounds had maximum values between 0.1 and 0.01 mg kg^{-1}, and the remaining 34 compounds had values between 0.01 and 0.001 mg kg^{-1}.

The total pesticide content in the soil ranged from 0.001 to 6.998 mg kg^{-1}, with a mean concentration of 0.104, and a median concentration of 0.016 mg kg^{-1} (the mean and median ranges refer to the positive samples only, i.e., samples with at least one pesticide ≥ LOQ). Most of the soil samples had relatively low pesticide content [58% of the samples had the total pesticide content below 0.05 mg kg^{-1}, 14% of the samples had the content between 0.05 and 0.15 mg kg^{-1}, and 7% of the samples had the content above 0.15 mg kg^{-1}; Figure 2]. The number of pesticide residues in soil samples and pesticide levels were poorly correlated with each other ($r^2 = 0.19$, $p < 0.01$; $r^2 = 0.28$, $p < 0.01$ if "NG_15" is not considered—this sample had 7 pesticide residues, total pesticide level: 6.998 mg kg^{-1}; Figure S1).

3.1.4. Predictors of Contamination. The number of residues in the soil was significantly correlated with the soil pH and soil organic carbon content (Table S5). Contaminated samples usually presented lower pH (average = 6.0, median = 6.0) and higher organic carbon content (average = 1.5, median = 1.1) than "pesticide-free samples" (pH average = 7.0, median = 7.6; OC average = 1.3, median = 1.2). The total pesticide content showed a significant and negative correlation with soil pH. As mentioned above, more significant correlations were found between the concentration of the most common pesticides and basic soil properties; a positive relationship was observed for organic carbon (atrazine-hydroxy-2), silt (AMPA), clay (metolachlor, atrazine-hydroxy-2, AMPA, and imidacloprid), and a negative relationship was observed for sand (atrazine-hydroxy-2 and AMPA).

The characteristics of the 63 pesticide residues found in the soil were highly variable, as were those of the 90 "tested and not found" residues. Nevertheless, some occurrence patterns/differences between the groups seem to exist: (i) 51% of the pesticide residues found in the soil are persistent (DT_{50} or $DT_{90} \geq 100$), yet a substantial amount of other persistent residues were not detected; (ii) 47% of the pesticide residues found in the soil present low solubility (<10 mg l^{-1}; low solubility residues represent 28% of the nondetected residues list); (iii) 70% of the residues found in the soil are moderate to highly hydrophobic (log $P \geq 2.7$; 50% in the nondetected list); (iv) 46% of the residues found in the soil are nonmobile or only slightly mobile in the soil (k_{oc} >500 mL g^{-1}; 37% in the nondetected list). There is no typical combination of characteristics among the most frequently detected compounds, with high persistence likely being the main driver of detection.

3.2. Country and Crop Profiles

The contaminated/clean sample ratio and the degree of contamination varied among countries and crop types (Figures 2−3). Most of the samples from Tunisia and South Sudan were "pesticide-free". Kenya also showed a relatively high proportion of pesticide-free samples (38%). On the other

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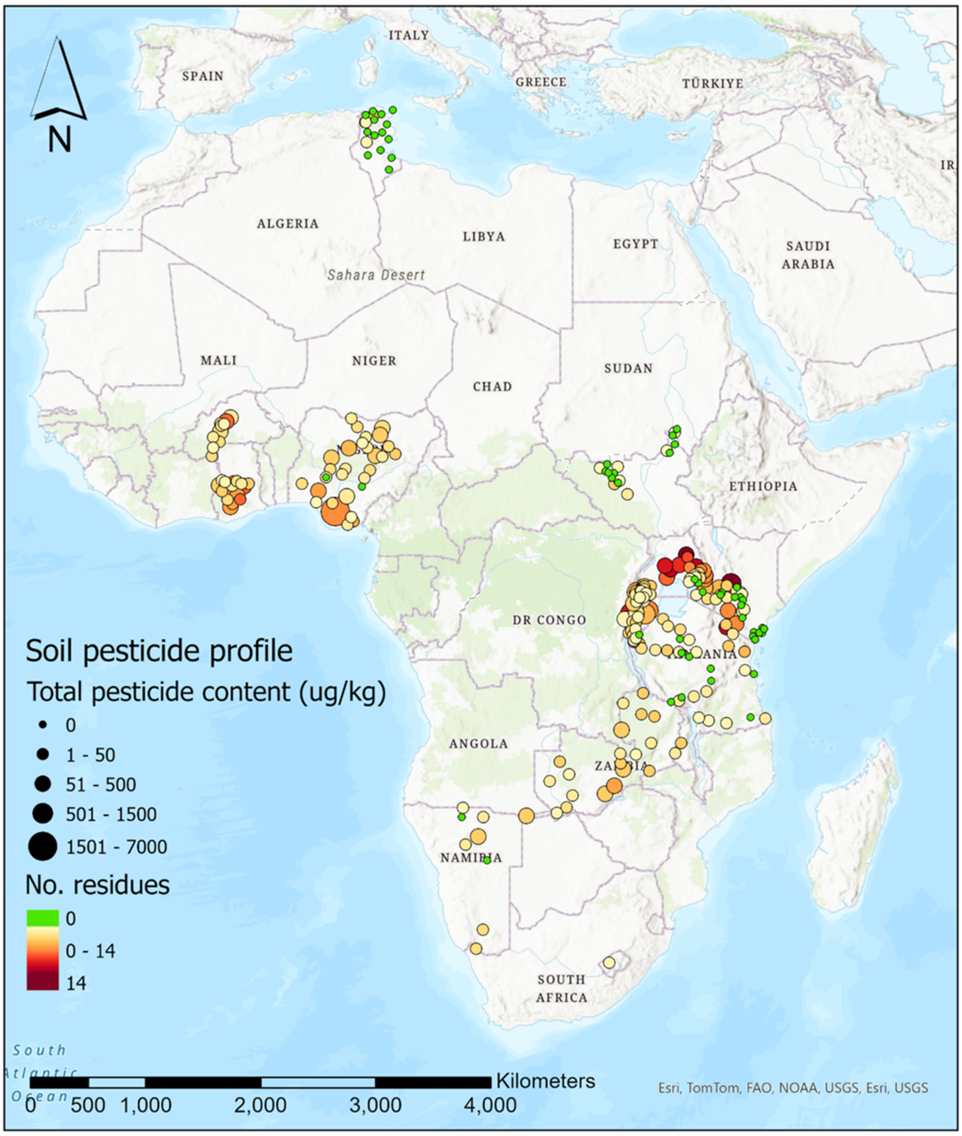

Figure 3. Overview of the number and total pesticide content in African agricultural samples.

hand, all soil samples from Burkina Faso, Ghana, Lesotho, Rwanda, Uganda, and Zambia (i.e., 6 out of the 13 countries covered) contained pesticide residues. The most common finding in these soils was a 2−5 residue mixture scenario. Ghana had the highest proportion of moderately complex mixtures (6−10 residues/sample), and Rwanda had the highest proportion of highly complex mixtures (>10 residues/sample). Note that our complexity interpretation here is only linked to the number of residues in the soil and not to the type of residues. Oilseed and other crops had a high percentage of "clean" soils, while, at the other extreme, all samples from the fields of fiber crops, fodder crops, and vegetables were contaminated. Cereals and seminatural areas, the classes with the highest sample representation, show a relatively similar pesticide profile to that of the overall data set (n = 272). Within these classes, there were samples with no pesticides, samples with a single residue, and samples with simple and more complex mixtures; simple mixtures were two to three times more common than the other contamination conditions (Figure 2).

The most complex mixtures, with 14 pesticide residues, were observed in Uganda and Kenya. Tanzania had a maximum of 12 residues/sample, Rwanda had 11, Burkina Faso and Ghana had 8, Nigeria had 7, Zambia had 6, and Burundi, Namibia, South Sudan, and Tunisia had a maximum of 4 pesticide residues/sample. The Lesotho samples had a single pesticide residue: metolachlor. The diversity of compounds and pesticide levels found in the soil adds complexity to contamination interpretation, and highlights national variances around pesticide use and/or persistence (see full national profiles in Supporting Information): 33 residues were found across Kenyan samples, 25 across Uganda samples, 20 in Ghana and Tanzania, 18 in Nigeria, 16 in Rwanda, 13 in Burkina Faso, 11 in Burundi, 10 in Namibia and Zambia, 6 in Tunisia, and 5 in South Sudan. The highest pesticide levels were found in the soils of Rwanda, Nigeria, and Uganda. Rwanda had a rather peculiar and high contamination level profile (see Figure S3 and Table S7 on significant differences, overall and cereal-specific). Soils from cereal fields and seminatural vegetation showed the most complex mixtures, with the highest number of residues per sample, and the

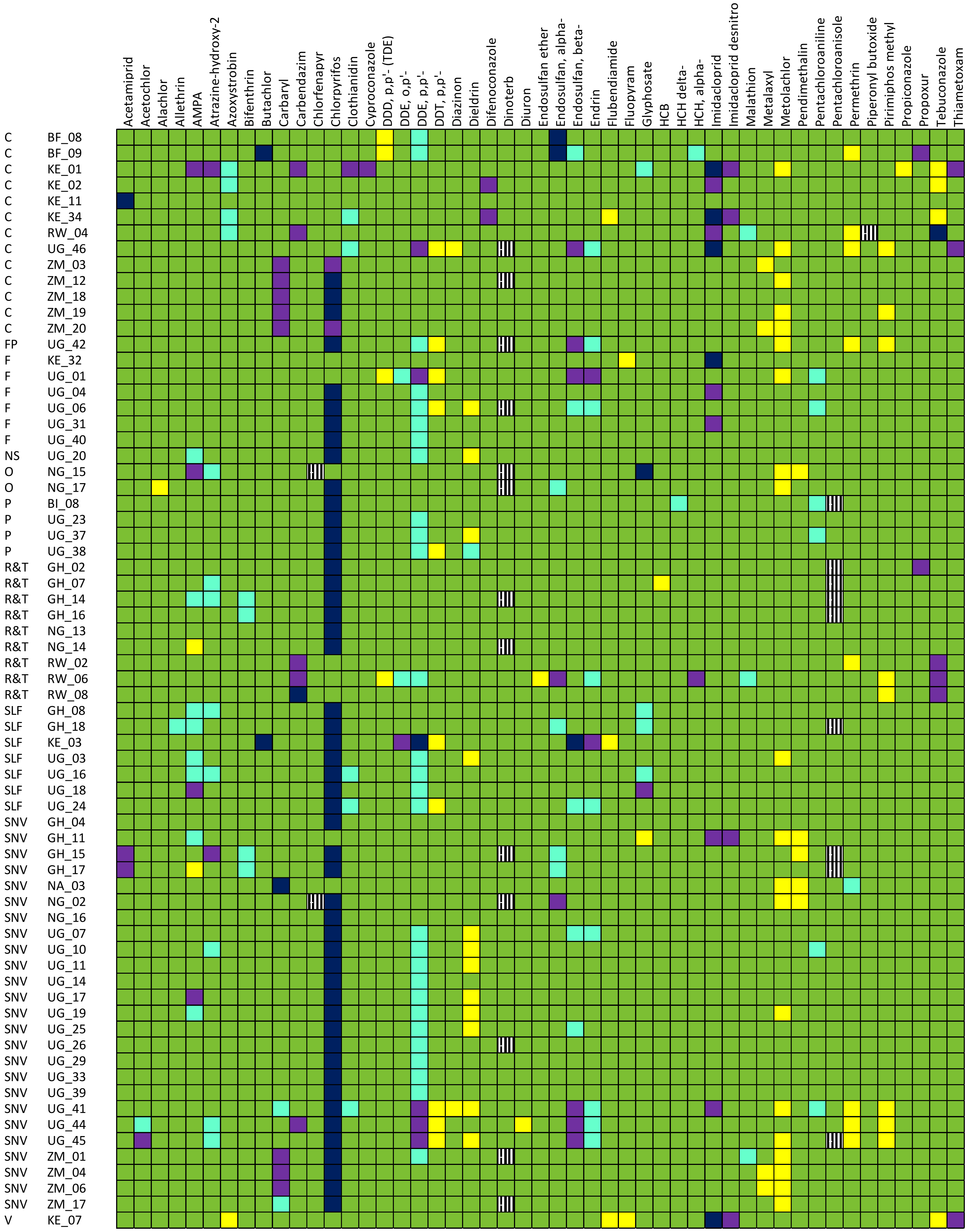

Figure 4. Matrix of the 69 soil samples with high-risk quotient ($\sum RQ \geq 1$). Samples are sorted by crop type (C = cereals; FP = fodder plants; *F* = fruits; NS = nursery stock; O = oil crops; *P* = pulses, R&*T* = roots & tubers; SLF = semiluxury foods; SNV = seminatural vegetation; V = vegetables), and pesticides are sorted alphabetically. Green cells represent cases where the compound was not found, white striped cells represent cases where the calculation of RQ was not possible, and yellow, light blue, purple, and dark blue represent cases of negligible, low, moderate, and

Figure 4. continued

high risk, respectively. Sample names are derived from country abbreviations as follows: Burkina Faso (BF), Burundi (BI), Ghana (GH), Kenya (KE), Lesotho (LS), Namibia (NA), Nigeria (NG), Rwanda (RW), South Sudan (SS), Tanzania (TZ), Tunisia (TN), Uganda (UG), and Zambia (ZM).

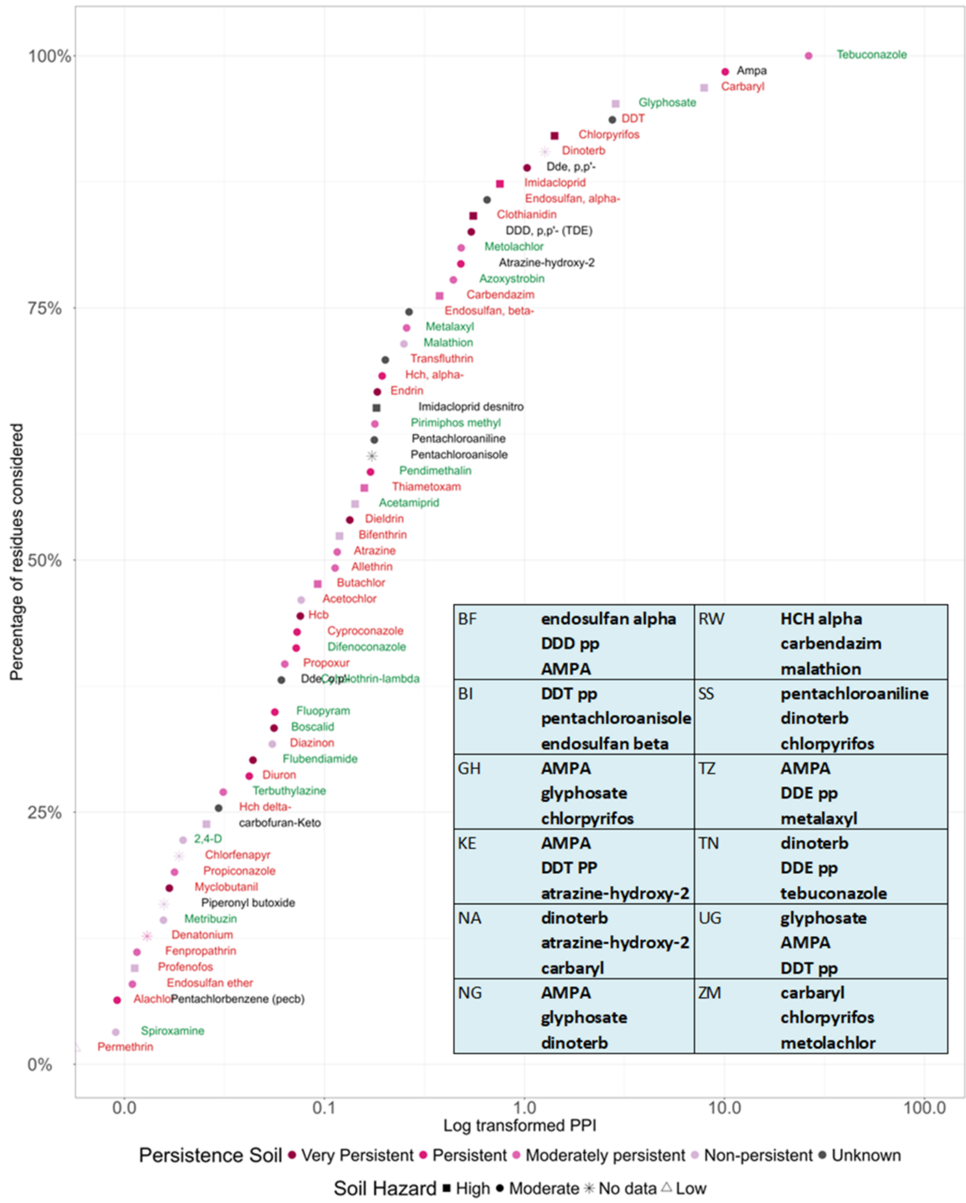

Figure 5. Pesticide prioritization indicator (PPI) curve, sorted by compounds from the lowest to highest PPI values (in log scale). Red, green, and black colors in the name of pesticide represent nonapproved (EU), approved, and metabolites, respectively. The light blue table covers the 3 compounds with the highest PPI values per country. See figure SM4 for country-specific PPI curves. The samples from Lesotho had only metolachlor. Country abbreviations: Burkina Faso (BF), Burundi (BI), Ghana (GH), Kenya (KE), Lesotho (LS), Namibia (NA), Nigeria (NG), Rwanda (RW), South Sudan (SS), Tanzania (TZ), Tunisia (TN), Uganda (UG), and Zambia (ZM).

highest diversity of compounds. Roots and tubers occupied the third position for both indicators. The highest levels in the soil were found in cereals, oil crops, and semiluxury foods (coffee, tea, cocoa, tobacco, nuts), and seminatural vegetation.

3.3. Environmental Risk Assessment

Sixty-nine samples/locations (25% of samples/locations assessed) showed an RQ ≥ 1 (high risk), 66 showed medium risk, 41 had low risk, 24 had negligible risk, and in 16 RQ could not be calculated (Figure 2). These 16 locations had one or a combination of the following compounds: chlorfenapyr, denatonium, dinoterb, pentachloroanisole, and piperonyl butoxide, for which no terrestrial ecotoxicity data were found. Note that these same data limitations may lead to the underestimation of risk in 75 more samples, where one or more of these missing data compounds appeared in a mixture with other compounds. In 66 of these samples, only one of these compounds was present in the mixture (often dinoterb), and in the remaining nine samples, two of these compounds

https://doi.org/10.1021/acs.est.5c15564
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were present in the mixture. Eight of these 75 samples/locations with incomplete RQ profiles are currently considered to be of negligible risk, 22 at low risk, 24 at moderate risk, and 21 of high risk. See Table S8 for possible underestimation cases. The analysis of risk quotients showed a clear difference across countries. The majority of samples from Uganda and Ghana were classified as high risk, followed by Zambia (45%, $n = 9$) and Rwanda (40%, $n = 4$). Burkina Faso, Nigeria, Kenya, Burundi, and Namibia showed high risk in 10−20% of samples, while no high-risk samples were observed in South Sudan, Lesotho, Tunisia, and Tanzania. In terms of crop types, the highest proportion of high-risk samples was observed in semiluxurious crops (54%, $n = 7$), followed by roots and tubers (45%, $n = 9$), and fruits (40%, $n = 6$). Nursery stock and vegetables, both with a very reduced number of samples, also had a high or even higher percentage of high-risk samples. Cereals, which represented the largest sampled group, had 12% of samples classified as high risk ($n = 13$), while 36% were in the medium-risk category ($n = 38$).

In 60 of the 69 high-risk samples, the exceedance of the moderate-high-risk threshold was due to a single pesticide (RQ ≥ 1). Most of the time, it was due to chlorpyrifos (RQ ≥ 1 in 50 samples); imidacloprid was responsible for the exceedance in 5 samples, while carbaryl, glyphosate, carbendazim, tebuconazole, acetamiprid, and endosulfan-α led to exceedance in one sample each. Only two samples contained more than one pesticide with RQ ≥ 1: BF_09 and KE_03. In the remaining seven samples, the high risk was due to the combination of multiple pesticides. Eleven pesticides had RQ values above 1. Besides the 8 compounds listed above, butachlor, DDE-p,p', and endosulfan-β have exceeded the high-risk threshold; most of these pesticides exceeded the threshold in only one or two samples (Figure 4).

3.4. Pesticide Prioritization Indicator (PPI)

Figure 5 represents the PPI plot when accounting for the median levels of pesticides across soil samples. The same exercise was done, but not presented for the 95th and 98th percentile pesticide levels. The top 11 compounds (in parallel with the 11 compounds that had RQ ≥ 1) in each of these curves were combined resulting in a 17 list compound: acetamiprid, AMPA, atrazine-hydroxy-2, butachlor, carbaryl, carbendazim, chlorpyrifos, clothianidin, DDE-p,p', DDT-p,p', dinoterb, endosulfan, α-, endosulfan, β-, glyphosate, imidacloprid, metolachlor, and tebuconazole. Five compounds appear in the RQ, PPI-50th, PPI-95th, and PPI-98th top 11 lists: carbaryl, chlorpyrifos, glyphosate, imidacloprid, and tebuconazole. DDE-p,p' and endosulfan-α appear in RQ and some of the PPI top lists. Acetamiprid, butachlor, carbendazim, and endosulfan-α emerge as top-priority compounds only under the RQ approach. This is because they are present in just a few samples, sometimes at extreme levels. PPI accounts for the frequency of detection and percentiles, which prevents these compounds from appearing in a top PPI position. AMPA, atrazine-hydroxy-2, clothianidin, DDT-p,p', dinoterb, and metolachlor appear exclusively in the PPI approach, owing to their widespread presence in soil or the limited availability of toxicological data; PPI calculation accounts for ecotoxicological data gaps. The top PPI/concern substances vary across countries. Some compounds, such as DDD-p,p', HCH-α, malathion, pentachloraniline, and pentachloroanisole, are overshadowed in the overall soil data set but stand out in a specific national context. AMPA ranked among the top concerns in six countries, dinoterb in four, glyphosate, DDT-p,p', and chlorpyrifos in three, and carbaryl, atrazine-hydroxy-2, and DDE-p,p' were top-ranked in two countries. The remaining eight compounds were top-ranked in only one country each (Figure 5). Country-specific PPI curves and related pesticide concern priorities are presented in Figure S4.

4. DISCUSSION

4.1. Unfolding Contamination Status

This study aimed to investigate the current contamination of African agricultural topsoils by pesticide residues, providing the first evidence of several compounds and their locations. The fact that the current study used international practices and standard operating procedures allowed its results to be compared to the European findings, such as those of Silva et al.,[15] Vieira et al.,[29] Knuth et al.,[30] and Rodríguez-Seijo et al.[31] Europe has the most explored and regulated system worldwide in terms of pesticides and soil contamination by pesticides. Overall, the African agricultural soil-pesticide profiles are close to the European ones in terms of the ratio of contaminated/uncontaminated samples (~80% contaminated soils) and the recurrent 2−5 mixtures output (~50−60 soils). Pesticide levels in African soils were, however, generally lower than those in Europe. The most recent European soil-pesticide data set by Knuth et al.,[30] which refers to midgrowing season samples, is expected to represent a worst-case scenario in terms of pesticide contamination. Pesticide levels reached values as high as 28 mg kg^{-1}, a value 4 times higher than our highest measurement in African soils. The lowest levels in African soils are most likely related to the lower pesticide application rates in Africa. The FAO estimates an average of 0.69 kg pesticide ha^{-1} in Africa, 1.64 kg ha^{-1} in Europe, and 2.37 kg ha^{-1} worldwide;[32] however, details on pesticide sales and use at a regional scale and/or applied or recommended application rates would be needed to validate this hypothesis of "lower soil levels due to lower application rates". Some information exists at the country level,[32] suggesting a relatively high intensity of pesticide use in Malawi, South Africa, and Rwanda. The first two countries were not covered in the current study, but our analyses of soils from Rwanda presented a high contamination profile. Other explanations for the low levels may be related to the sampling time. Low levels may reflect just background contamination if sampling was performed before the growing season. Although we have the sampling dates of the soils and an overall understanding of the typical growing season timeline of the main crops, the lack of proper management records or validation of our assumptions hampers further temporal analyses. The high temperatures in most countries and the low organic matter content of most of the soil samples may be relevant factors too, limiting soil adsorption potential and storage capacity for pesticides.[33] Other factors possibly contributing to lower contaminant levels in African soils include the generally high ultraviolet (UV) radiation in African regions, which can enhance photolytic degradation of some pesticides,[34] and the near absence of snow cover, which limits atmospheric scavenging and deposition of volatile compounds.[35−37] Further exploring the influence of soil characteristics, including microbial community distribution, and climate factors on the fate and degradation/persistence of pesticides via laboratory, modeling, and statistical frameworks, could open the possibility of using soil-climate indicators as predictors of contamination. See Riedo et al.,[38] Vinther et

https://doi.org/10.1021/acs.est.5c15564
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al.,[39] and Chakraborty and Ebihara[40] for insights on the possible influence of geoclimatic conditions on pesticide dynamics. The establishment and validation of such predictors would allow more targeted monitoring programs (given the high cost of pesticide residue analyses in soils, it may be necessary not only to prioritize which pesticides to analyze but also to identify which soils/sites should be sampled and analyzed), higher environmental protection (via earlier interventions and more informed practices), and prioritizing areas for detailed investigations and remediation actions.

4.2. Most Relevant Pesticides

The list of most common pesticides in African soils—dinoterb, DDE-p,p', metolachlor, chlorpyrifos, AMPA, atrazine-hydroxy-2, and imidacloprid—highlights the high use and/or persistence of herbicides and insecticides, current and historical drivers of contamination, relevance of metabolites in soil mixtures, and the "omnipresence" of highly hazardous compounds in agricultural settings. Chlorpyrifos and imidacloprid have been flagged by previous African studies.[4] European studies corroborate some of them as frequent soil contaminants: DDE-p,p′, AMPA, chlorpyrifos, alongside HCB, and glyphosate.[30] The review by Sabzevari and Hofman,[4] and our literature search (not restricted to currently used pesticides), highlighted some other compounds as frequent in African soils. Such a mismatch with our measurements suggests a very localized use of some pesticides or recent changes in pesticide use. Dimethoate, dichlorvos, and mevinphos, for instance, were reported to be highly frequent in North Nigeria,[41] but were not found in our soil samples. Another possible explanation, at least for these compounds, all of which are nonpersistent in soil, relates again to soil sampling time, which in our case was not "controlled". The pesticide soil samples were embedded in the overall Soils4Africa sampling campaign and may have fallen outside of the growing season or too long after the application of some nonpersistent compounds to capture them. To the best of our knowledge, only a couple of studies have explored the oscillations of pesticide-soil levels over the year. Honert et al.[42] and Veloso et al.[43] showed that mixtures of pesticide residues can be found all year-round, but significant changes in the number, levels, and composition of the mixtures occur over the year.

The levels of these common pesticides and the total pesticide content were overall considered low, especially in relation to the European values. The interpretation of pesticide findings is complex and limited here and in general. This occurs mostly because of the lack of quality thresholds and oil screening values (SSV). Different methods are used to derive SSV (see, e.g., the EEA[44] suggestion on using agricultural critical limits, and the four soil data-based methods proposed by the European Joint Program on Soil (EJP SOIL[45])), as the number of SSV values for pesticides is very limited (see, however, the detailed work of ConSOIL in this regard[46]). Comparison with levels in African studies could inform trends of contamination or even the use of individual substances; however, the different sampling periods, different sampling strategies, different analytical methods, and different analyte lists among studies hamper such detailed analyses. Moreover, the lack of detailed pesticide use information hampered comparisons between predicted and measured environmental concentrations, which is a practical and fast way to infer off-site transport of pesticides and degradation/accumulation dynamics. See the discussion by Knuth et al.[30] on PEC-MEC comparisons and mismatches in the European context. Theoretical risk assessments via quotient analyses based on the hazard profile of pesticides in soil are a common output and complement occurrence and level assessments.[27] Our results showed that Ghana and Uganda had the highest proportion of high-risk samples, but little can be said in this regard. Regional inferences require access to pesticide use records and pesticide use and risk reduction efforts, which, unfortunately, are often not available or centralized. The high-risk sample/case output was primarily driven by the presence of chlorpyrifos. Chlorpyrifos is frequently detected in soils worldwide,[4] and is a main contributor to risk, at least in Nepal[47] and Europe.[48] Note, however, the uncertainty and implications of missing ecotoxicity data for some pesticides. Dinoterb is of particular concern because of its relatively high concentration and frequent detection across samples. The Pesticide Prioritization Indicator, by accounting for toxicity data gaps, delivers more sensitive and conservative rankings. Yet, as no risk or urgency threshold exists in this regard, a combination with other approaches is needed. The application of more comprehensive modeling approaches and the performance of a battery of ecotoxicological assays with realistic mixtures could be an important step forward toward the characterization of the health of African soils and the risks associated with African agricultural practices.

4.3. Strengths and Limitations

In our opinion, this study may serve as a soil-pesticide contamination baseline for the African continent and a key indicator of soil quality and soil health assessments. Our results can be used as a first soil contamination indication and serve as a reference in some locations (e.g., Burundi, South Sudan), and they can support simple contamination trend analyses in more documented ones (e.g., Ghana, Nigeria, Kenya, Tanzania). The number of samples in this study is rather low to derive (sound) conclusions at the African continent level, but may still be sufficient to provide some guidelines in terms of research agenda and policy recommendations, even at the global scale. The high frequency of non-EU-approved substances is closely linked to discussions and petitions against exporting EU-banned pesticides. The exercise done with RQ and PPI is expected to help set regulatory (re-evaluation) priorities and, altogether, support the design of pesticide reduction strategies. See Wan et al.[49] and Brunelle et al.[50] on what more sustainable production may entail from a more holistic lens, and Schwirn et al.[51] on the crucial role of innovation and legislation in such a journey. See Zhang and Makowski,[52] Ediagbonya et al.,[53] Ullah et al.,[54] Deguine et al.,[55] and Wan et al.[56] on the performance of different pesticide reduction strategies (e.g., integrated pest management, biological control, crop diversification, precision agriculture, cocultures of crops, and domestic animals), and Bakker et al.[57] and Clausen et al.[58] on the impact of risk perception of pesticides and capacity building of farmers in the adoption and success of pesticide reduction plans. From a more fundamental research perspective, our data set can also be valuable to explore the definition of pesticide thresholds in soil, or, for example, to corroborate the co-occurrence of contaminants (pesticides plus plastics/heavy metals/etc.), which is a crucial aspect for field-scale processes.[59]

The mismatch between the compounds found in soil and those registered for use in African countries suggests legacy effects, transboundary contamination, and/or illegal use of

https://doi.org/10.1021/acs.est.5c15564
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some pesticides. Such a mismatch underscores the importance of comprehensive application records, the relevance of the country and sample context, and the need for a structured framework for identifying the sources or causes of non-approved substances in soils. The double-entry tables proposed by Chiaia-Hernandez et al.,[60] which link applied and detected residues in soil, the rationale outlined by Riedo et al.[1,38] regarding pesticides in organic soils, and Benzing et al.[61] decision tree for pesticide residues in organic food offer valuable guidance for developing such an evaluation framework. Dinoterb is an especially intriguing case. The monitoring results suggest widespread use, but it was not found in any national (agricultural) registration database or list. Some registered compound lists were retrieved from reports published prior to soil sampling. It is possible that new substances have been introduced in the market since this report was published. Other lists were obtained from databases that may have been updated after the sampling period and may not reflect compounds registered at the time but discontinued thereafter. Having information on not approved and the expiration dates of approval status, like in the EU pesticide database, could also be important to clarify some findings. More insights into the sources of the not registered pesticides could come from further information on local/regional/international pesticide use (including outside agricultural settings; dinoterb, for instance, seems to be used as a finishing substance for pest control in textiles),[62] and regulatory use exemptions/derogations.

The biggest limitation of this study relates to the nonpesticide-focused aspect of the Soils4Africa soil campaign. Adjustments were made to the default project procedures to support this study (soil depth and storage conditions were adjusted), but challenges around the project field campaign led to a long sample collection period and sometimes a large window between sampling and samples arriving at the analytical facilities. This situation may be confounding and may dilute some conclusions. Others may be hampered by the reduced and uneven numbers of samples in some countries, regions, and crop types. A higher sample coverage improves the representativity of the data set and also helps to better understand the fate and impacts of pesticides in an agricultural context and beyond. This is highly relevant considering the local site characteristics; most of the pesticide sample locations show erosion signs, and there is growing evidence that pesticides contaminate on a landscape scale, far beyond the fields.[28,63] Having more information about yields, pesticide practices, and pesticide-related programs before sampling could have led to a greater impact of this research on farmers and policymakers. Having such information could even allow the identification of contamination hotspots, i.e., areas of high pesticide use/high contamination with pesticides, where assessing and addressing soil health should be the most urgent. We identified some during the field campaign, namely, in Uganda (which may explain the presence of four UG samples among the small group of samples with more than ten pesticide residues), but it was not possible to pursue this effort further considering the low return response of our questionnaire, how challenging and time-consuming the identification of hotspots in the field was, and the reservations of landowners to collect samples in these areas. The analytical data set identified other hotspots, such as NG_15. It is, therefore, rather urgent that these areas be tackled to ensure that soil monitoring plans, similar to those that will be implemented due to the recently adopted Soil Monitoring Law in Europe,[64] do not underestimate true soil conditions.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c15564.

Analytical data set (Table S2; also in ZENODO, Wageningen University & Research (2025). Soils4Africa - Pesticide dataset (Version v01) [Data set]. Zenodo. https://doi.org/10.5281/zenodo.177779527), pesticide characteristics (Table S3), and the hazard database for the 63 pesticides found in the soil (Table S4) (XLSX)

Figures S1−S4; tables on the sample distribution country-crop class, correlation matrix between the occurrence of pesticide residues, and basic soil properties; profile of the most common pesticide residues in soil; Risk Quotient uncertainty analyses; and country and crop pesticide profile (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was funded by the H2020-SFS-2019-2 grant agreement No. 862900 (Soils4Africa) from the European Union's Horizon 2020 Research and Innovation Program and was supported by grant agreement No. 857560 (CETOCOEN Excellence). This publication reflects only the author's views, and the European Commission is not responsible for any use that may be made of the information it contains. The authors would like to thank the field coordinators and everyone involved in the pesticide field campaign, Hans Mol, and Paul Zommer from Wageningen Food Safety Research, for their support in analytical work and interpretation. The authors also thank the RECETOX Research Infrastructure (No LM2023069), financed by the Ministry of Education, Youth and Sports, for the supportive background.

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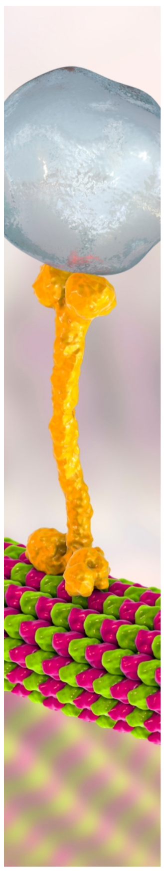

https://doi.org/10.1021/acs.est.5c15564
Environ. Sci. Technol. 2026, 60, 17446−17460