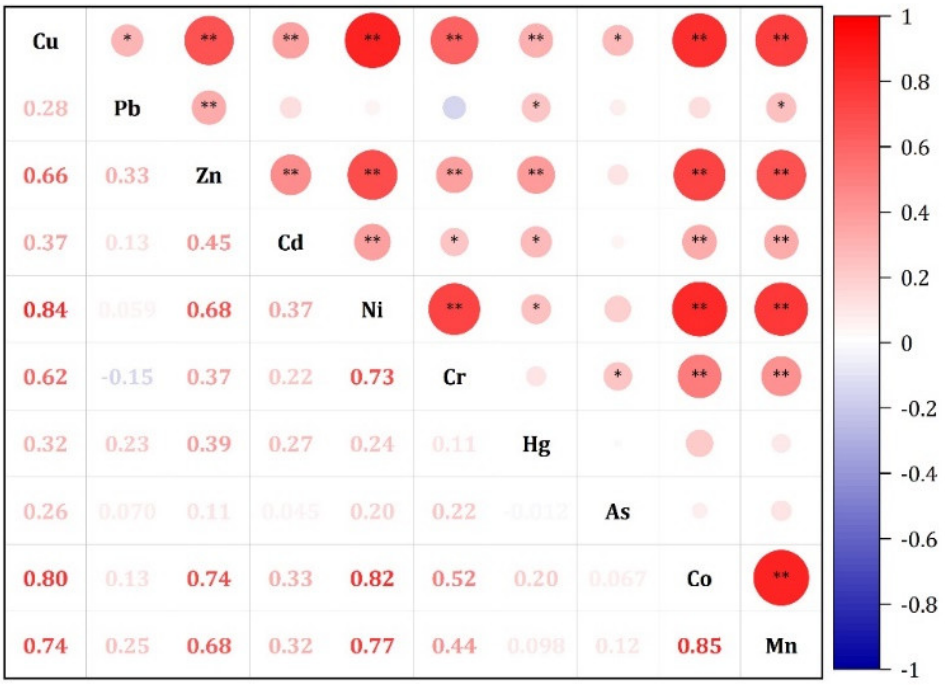


**Figure S1.** Pearson correlation analysis of HMs in soil.

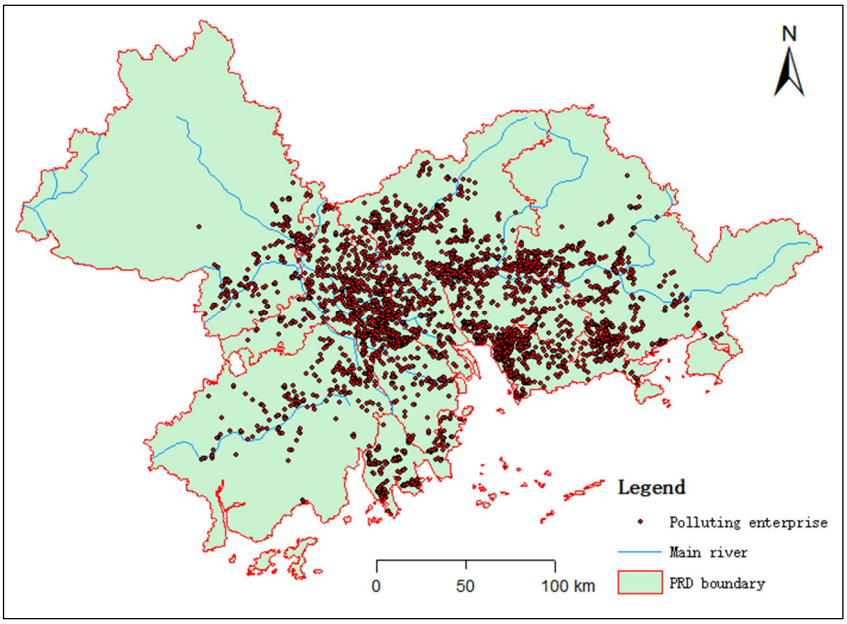


**Figure S2.** Spatial distribution of 1904 enterprises associated with surface treatment of metals and manufacturing in PRD.

**Table S1.** Statistic description information of quality control of soil HMs.

| HMs | Parallel samples | | Laboratory controlling samples | | Minimum detection Limit |
|---|---|---|---|---|---|
| | Absolute deviation % | Qualification rate % | Recovery rate % | Qualification rate % | mg/kg |
| Cu | 0–9.1 | 100 | 90–102 | 100 | 0.003 |
| Pb | 0.8–13.5 | 100 | 87–110 | 100 | 0.001 |
| Zn | 0.4–10.0 | 100 | 90–120 | 100 | 0.003 |
| Cd | 0–6.7 | 100 | 84–110 | 100 | 0.0015 |
| Ni | 0–18.0 | 100 | 80–120 | 100 | 0.0025 |
| Cr | 0–10.0 | 100 | 84–111 | 100 | 0.01 |
| Hg | 0.4–7.1 | 100 | 88–106 | 100 | 0.002 |
| As | 0.3–1.4 | 100 | 92–105 | 100 | 0.003 |
| Co | 0.3–9.0 | 100 | 90–103 | 100 | 0.005 |
| Mn | 0.3–9.0 | 100 | 84–104 | 100 | 0.001 |
| pH | 0.01–0.27 | 100 | / | / | / |

**Table S2.** Geo-accumulation Index and pollution levels.

| Contamination level | $I_{geo}$ Value |
|---|---|
| Unpolluted | $I_{geo} \leq 0$ |
| Unpolluted to moderately polluted | $0 < I_{geo} \leq 1$ |
| Moderately polluted | $1 < I_{geo} \leq 2$ |
| Moderately to heavily polluted | $2 < I_{geo} \leq 3$ |
| Heavily polluted | $3 < I_{geo} \leq 4$ |
| Heavily to extremely polluted | $4 < I_{geo} \leq 5$ |
| Extremely polluted | $I_{geo} > 5$ |

**Table S3.** Total variance of HMs explained by PCA.

| Component | Initial eigenvalues | | | Extraction sums of squared loadings | | | Rotation sums of squared loadings | | |
|---|---|---|---|---|---|---|---|---|---|
| | Total | % of variance | Cumulative% | Total | % of variance | Cumulative% | Total | % of variance | Cumulative% |
| 1 | 4.847 | 48.468 | 48.868 | 4.847 | 48.468 | 48.468 | 4.426 | 44.264 | 44.264 |
| 2 | 1.369 | 13.695 | 62.163 | 1.369 | 13.695 | 62.163 | 1.743 | 17.429 | 61.693 |
| 3 | 1.015 | 10.153 | 72.316 | 1.015 | 10.153 | 72.316 | 1.062 | 10.623 | 72.316 |
| 4 | 0.922 | 9.222 | 81.537 | | | | | | |
| 5 | 0.724 | 7.239 | 88.777 | | | | | | |
| 6 | 0.466 | 4.665 | 93.441 | | | | | | |
| 7 | 0.279 | 2.787 | 96.229 | | | | | | |
| 8 | 0.167 | 1.669 | 97.897 | | | | | | |
| 9 | 0.120 | 1.196 | 99.093 | | | | | | |
| 10 | 0.091 | 0.907 | 100.000 | | | | | | |

**Table S4.** Matrix of principal components analysis.

| Component | Component matrix | | | Rotated component matrix | | |
|---|---|---|---|---|---|---|
| | PC1 | PC2 | PC3 | PC1 | PC2 | PC3 |
| Cu | 0.915 | −0.031 | 0.113 | **0.851** | 0.294 | 0.200 |
| Pb | 0.254 | 0.721 | 0.398 | −0.045 | **0.821** | 0.261 |
| Zn | 0.831 | 0.268 | −0.044 | **0.704** | **0.518** | −0.023 |
| Cd | 0.497 | 0.284 | −0.171 | **0.405** | **0.400** | −0.181 |
| Ni | 0.922 | −0.228 | −0.061 | **0.946** | 0.084 | 0.072 |
| Cr | 0.661 | −0.511 | −0.023 | **0.782** | −0.259 | 0.144 |
| Hg | 0.356 | 0.559 | −0.085 | 0.175 | **0.623** | −0.167 |
| As | 0.223 | −0.246 | 0.880 | 0.150 | −0.007 | **0.929** |
| Co | 0.900 | −0.086 | −0.158 | **0.895** | 0.193 | −0.054 |
| Mn | 0.856 | −0.044 | −0.042 | **0.824** | 0.237 | 0.046 |

Note: significant loading factors are remarked in bold.

## Calculation of source apportionment contribution by PCA-MLR

**Table S5**. Contribution composition of soil HMs in PRD

| $B_i$ | $B_i / \sum B_i$ | contribution ratios |
|---|---|---|
| 0.864 | 0.643336 | 64.33% |
| 0.333 | 0.247952 | 24.80% |
| 0.146 | 0.108712 | 10.87% |

**Table S6.** The factor score variables of source $i$ at 80 sampling site

| $FS_1$ | $FS_2$ | $FS_3$ |
|---|---|---|
| 1.59935 | −0.97522 | −0.57611 |
| −0.58927 | −0.17569 | −0.13999 |
| 0.35888 | −0.91572 | −0.01049 |
| −0.63309 | −0.03203 | −0.2231 |
| −0.56855 | −0.41329 | −0.23399 |
| 0.01301 | 0.02439 | −0.36446 |
| 0.23303 | −1.18341 | 0.5364 |
| −0.21159 | −0.55683 | −0.47919 |
| −0.73656 | −0.388 | −0.20532 |
| −0.68157 | −0.68829 | −0.36507 |
| −0.70573 | −0.83749 | −0.33256 |
| 0.20352 | −0.22494 | −0.27362 |
| −0.74366 | −0.69149 | −0.10914 |
| 0.41419 | −0.04575 | 0.25792 |
| 0.3352 | 0.02871 | −0.44512 |
| −0.19295 | −0.96929 | −0.67459 |
| 3.10929 | −0.30915 | −0.53833 |
| 0.6943 | −2.13327 | 0.10467 |
| 0.68871 | 2.82774 | −0.81545 |
| 0.37005 | 0.23339 | −0.11192 |
| 0.34248 | 0.80104 | −0.62255 |
| 0.68983 | 0.68155 | 1.11868 |
| −0.08633 | −0.58187 | −0.42384 |
| −0.3401 | 0.23472 | −0.34013 |

| | | |
|---|---|---|
| −0.19969 | 0.16781 | −0.48361 |
| −0.01385 | 1.52021 | −0.52085 |
| −0.22321 | −0.45298 | −0.86111 |
| −0.40772 | −0.59537 | −0.79129 |
| −0.55867 | 0.20155 | −0.05595 |
| −0.25283 | −0.74214 | 0.85345 |
| −0.14843 | 1.00515 | 0.06663 |
| −0.96479 | 0.33147 | 0.17081 |
| −0.34109 | 0.10276 | −0.64414 |
| 2.86174 | 0.23253 | 0.08164 |
| −0.83498 | 0.25237 | −0.45145 |
| 0.44031 | −0.94878 | 0.98972 |
| 2.60409 | 0.17252 | −0.19999 |
| −0.13281 | −0.41996 | −0.25429 |
| 2.32914 | 0.15039 | 0.53142 |
| −0.40922 | 1.64504 | −0.30789 |
| −1.5615 | 3.51419 | 2.85046 |
| 2.3062 | 0.05777 | 0.96728 |
| 3.57539 | 0.26188 | 0.54861 |
| 2.52806 | 2.51712 | 0.4644 |
| −0.20695 | −0.61737 | −0.36651 |
| −0.38455 | 1.85715 | −0.5131 |
| 0.50115 | 0.32955 | 0.39736 |
| −0.40594 | 1.51915 | −2.0581 |
| −0.20468 | 0.26051 | −0.03479 |
| −0.235 | 0.29476 | −0.16864 |
| −0.50863 | −0.02206 | −0.20607 |
| 0.13159 | −0.51676 | −0.31867 |
| −0.26074 | −1.18914 | −0.48439 |
| −0.08144 | −0.4937 | 0.73001 |
| −0.38685 | −0.21076 | −0.37915 |
| −0.30387 | 1.4737 | −0.68755 |
| −0.04721 | 0.84838 | −0.0184 |
| −0.70794 | −0.64628 | −0.64502 |
| −0.10908 | −0.69738 | −0.02907 |
| −0.11241 | −0.80925 | 0.6144 |
| −0.45091 | −0.46058 | −0.26724 |
| −0.74535 | 0.38924 | −0.7628 |
| −0.7611 | −0.78822 | −0.84504 |
| −0.06666 | −1.31658 | −0.59305 |
| −0.10908 | −0.69738 | −0.02907 |
| −0.69977 | 1.15503 | −0.6432 |
| −1.43805 | 2.74193 | 0.71731 |
| −0.20495 | −1.02519 | −0.00028 |
| −0.25031 | −0.15655 | 0.1785 |
| −0.24868 | −1.09505 | 5.31293 |
| −0.43242 | 0.25542 | −0.61207 |
| 0.33792 | 0.41083 | 0.40517 |
| 0.0292 | −1.63361 | −0.33177 |
| −0.89145 | 0.1431 | −0.27243 |
| −0.92195 | −0.24492 | 0.53467 |
| −0.676 | −0.05019 | −0.56624 |
| −1.0524 | −0.45116 | 4.43639 |

| | | |
|---|---|---|
| −0.80491 | −0.67332 | −0.11805 |
| −0.84303 | −0.37113 | −0.53552 |
| −0.60614 | −0.1955 | 0.4729 |

**Table S7.** Contributions of four sources in 80 sampling site

| Site | PC1 | PC2 | PC3 |
|---|---|---|---|
| 1 | 156.7625 | −13.5 | −0.89489 |
| 2 | −6.27614 | 9.4554284 | 4.595017 |
| 3 | 64.35515 | −11.79169 | 6.225173 |
| 4 | −9.54046 | 13.580072 | 3.548823 |
| 5 | −4.73263 | 2.6336588 | 3.411739 |
| 6 | 38.58998 | 15.199956 | 1.769373 |
| 7 | 54.98011 | −19.47737 | 13.10946 |
| 8 | 21.85868 | −1.48754 | 0.325143 |
| 9 | −17.2483 | 3.3597638 | 3.772639 |
| 10 | −13.1519 | −5.261908 | 1.761694 |
| 11 | −14.9517 | −9.545612 | 2.170932 |
| 12 | 52.7818 | 8.0414042 | 2.912873 |
| 13 | −17.7772 | −5.353784 | 4.983359 |
| 14 | 68.47541 | 13.186155 | 9.603937 |
| 15 | 62.59114 | 15.323988 | 0.754019 |
| 16 | 23.24724 | −13.32974 | −2.13457 |
| 17 | 269.2436 | 5.6236381 | −0.41932 |
| 18 | 89.34186 | −46.74895 | 7.674815 |
| 19 | 88.92544 | 95.687362 | −3.90772 |
| 20 | 65.18725 | 21.200586 | 4.948365 |
| 21 | 63.13346 | 37.498471 | −1.47948 |
| 22 | 89.00888 | 34.067776 | 20.43925 |
| 23 | 31.18977 | −2.206467 | 1.021893 |
| 24 | 12.28548 | 21.238772 | 2.075641 |
| 25 | 22.74515 | 19.317709 | 0.269504 |
| 26 | 36.58908 | 58.14667 | −0.19928 |
| 27 | 20.99306 | 1.4941132 | −4.48249 |
| 28 | 7.248207 | −2.594068 | −3.60359 |
| 29 | −3.99663 | 20.286423 | 5.652919 |
| 30 | 18.78655 | −6.808003 | 17.10051 |
| 31 | 26.5637 | 43.358705 | 7.195965 |
| 32 | −34.2501 | 24.016576 | 8.50739 |
| 33 | 12.21173 | 17.450049 | −1.75126 |
| 34 | 250.8027 | 21.175895 | 7.384912 |
| 35 | −24.58 | 21.745524 | 0.674336 |
| 36 | 70.42118 | −12.74088 | 18.81589 |
| 37 | 231.6094 | 19.452938 | 3.839733 |
| 38 | 27.7273 | 2.4421554 | 3.156201 |
| 39 | 211.1273 | 18.817561 | 13.04678 |
| 40 | 7.136467 | 61.730683 | 2.48148 |
| 41 | −78.7012 | 115.39613 | 42.23902 |
| 42 | 209.4184 | 16.158334 | 18.53341 |
| 43 | 303.9652 | 22.018567 | 13.26316 |
| 44 | 225.9456 | 86.769105 | 12.20312 |

| | | | |
|---|---|---|---|
| 45 | 22.20433 | −3.225713 | 1.743567 |
| 46 | 8.974229 | 67.820606 | −0.10172 |
| 47 | 74.95339 | 23.961451 | 11.35922 |
| 48 | 7.380806 | 58.116237 | −19.5503 |
| 49 | 22.37343 | 21.979233 | 5.919283 |
| 50 | 20.11478 | 22.96259 | 4.234369 |
| 51 | −0.26896 | 13.866323 | 3.763198 |
| 52 | 47.42346 | −0.337084 | 2.345781 |
| 53 | 18.19731 | −19.64189 | 0.259685 |
| 54 | 31.55404 | 0.3249951 | 15.54664 |
| 55 | 8.802893 | 8.4485283 | 1.584454 |
| 56 | 14.98439 | 56.811315 | −2.29771 |
| 57 | 34.10397 | 38.857657 | 6.125601 |
| 58 | −15.1163 | −4.055752 | −1.76234 |
| 59 | 29.49504 | −5.522892 | 5.991286 |
| 60 | 29.24697 | −8.734809 | 14.09133 |
| 61 | 4.03082 | 1.2759084 | 2.993185 |
| 62 | −17.9031 | 25.675219 | −3.24496 |
| 63 | −19.0764 | −8.131013 | −4.2802 |
| 64 | 32.65506 | −23.30084 | −1.10813 |
| 65 | 29.49504 | −5.522892 | 5.991286 |
| 66 | −14.5077 | 47.661932 | −1.73943 |
| 67 | −69.505 | 93.223658 | 15.38677 |
| 68 | 22.35331 | −14.93469 | 6.353697 |
| 69 | 18.97428 | 10.00496 | 8.604193 |
| 70 | 19.0957 | −16.94046 | 73.23677 |
| 71 | 5.40821 | 21.833093 | −1.34756 |
| 72 | 62.79376 | 26.295093 | 11.45753 |
| 73 | 39.79604 | −32.40313 | 2.180877 |
| 74 | −28.7867 | 18.608256 | 2.927853 |
| 75 | −31.0587 | 7.4677554 | 13.08769 |
| 76 | −12.737 | 13.058678 | −0.77065 |
| 77 | −40.7765 | 1.5463675 | 62.20282 |
| 78 | −22.34 | −4.832102 | 4.8712 |
| 79 | −25.1797 | 3.8441209 | −0.38394 |
| 80 | −7.53285 | 8.8866605 | 12.31012 |