Fig. S1  SIRM 77 K (10^{-6} \text{ Am}^2 \text{ kg}^{-1}) and estimated magnetite concentration (\mu g \text{ g}^{-1}) for frontal cortex samples versus age at death, Mexico City and Manchester cases. The annual mean airborne PM$_{2.5}$ concentration (\mu g m^{-3}) is given for the residence area of the Mexican cases (inside each data symbol); SIRM values for gray (g) and white (w) matter are given for the Manchester cases, together with their clinical diagnosis upon death (CAA = cerebral amyloid angiopathy; CVD = cerebrovascular disease; DLB = dementia with Lewy bodies, see Table 1).
Fig. S2: Magnetic analyses of brain tissue samples (freeze-dried): A) acquisition of isothermal (RT) remanent magnetization in applied direct current (DC) fields from 5 mT to 1 T. All samples acquire most of their magnetization at fields < 100 mT, indicating the dominant presence of ferrimagnetic minerals (e.g. magnetite and/or maghemite). The magnetically-softest sample (the Mexico City case to the left of all remaining samples) has the highest SIRM value (case 282). B) Measurement of LT remanence (77 K, dc field 1 T) upon warming to RT, showing the thermal unblocking of the superparamagnetic particles. C) Comparison between the brain samples and sized, synthetic magnetites of known grain size and degree of dispersion\(^{30}\), as measured by the RT anhysteretic remanent magnetization (ARM), normalized by the SIRM, plotted against the median destructive field of the ARM (MDF\(_{\text{ARM}}\), in mT). All of the measurable brain samples fall within the region of the least-dispersed synthetic, sub-micrometre magnetites, indicating magnetic interactions, and hence agglomeration/clustering of some of the brain magnetite particles.
**Fig. S3:** High-angle annular diffraction (HAADF) (a) and dark-field (b) TEM micrographs showing spherical magnetic nanoparticles in brain tissues. (c) Fe-L$_{2,3}$ EELS spectra of nanoparticles identified in the selected areas (boxes: 1-4) showing the absence of any pre-edges (see hematite, goethite and ferrihydrite pre-edge at ~708.8 eV), Fe-L$_3$ edges centered at 708 eV and broad Fe-L$_2$ features characteristic of magnetite, compared with the Fe-L$_{2,3}$ EELS spectra in (d) of standard magnetite, siderite, hematite, goethite and 2-line-ferrihydrite.
**Fig. S4:** Fe-L$_{2,3}$-edge spectra of magnetic particles found in brain samples. The Fe-L$_3$ and Fe-L$_2$ edges in all three samples are at 708.7-709.8 and 72-723 eV, in excellent agreement with the chemical shift in EELS spectra for the magnetite structure (also see Fig. S3).
**Fig. S5**: Particle size distribution of magnetic particles in brain magnetic extracts. Particle size measurements were carried out on all the HRTEM micrographs collected from 6 brain magnetic extracts from different subjects. The ImageJ software package was used to describe the imaged particles (spherical and non-spherical) in terms of the longest and shortest diameters, perimeter projected area, or equivalent spherical diameter.
Fig. S6: Co-L_{2,3} EELS spectra of cobalt (II,III) oxide nanoparticles associated with magnetite particles in brain tissues. Co-L$_3$ and Co-L$_2$ edges from different areas of a brain tissue sample (a) are centred at ~780 and ~796 eV, respectively, in a good match with an EELS spectrum of a standard cobalt (II,III) oxide.
**Fig. S7:** Energy dispersive X ray analysis of metal-bearing NPs in brain tissue samples, showing presence of Fe, Ni, Co (and possibly Cu, with the caveat that the samples were mounted on holey carbon films on Cu grids).
**Fig. S8:** Energy dispersive X ray analysis of metal-bearing NPs in brain tissue samples, showing the presence of Fe, Ni, Pt, Co and possibly Cu.
Fig. S9: A collection of HRTEM micrographs of magnetite particles, extracted from brain tissues, showing dominant rounded morphologies. Micrograph (c) shows fused magnetite particles and micrographs (d) and (e) show aggregated magnetite particles.
**Fig. S10:** HRTEM micrograph of magnetically-extracted magnetite particles from brain tissues. (b-d) FFT patterns of selected areas (1-3, respectively) featuring a single crystal in (b) and magnetite particles superimposed at ~90° in (c) and (d).
**Fig. S11:** TEM image of magnetite nanoparticles captured from the exhaust plume of a diesel engine (adapted from Liati et al., 2015).