Magnetic properties of atmospheric particulate matter from automatic air sampler stations in Latium (Italy): Toward a definition of magnetic fingerprints for natural and anthropogenic PM_{10} sources

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[1] Environmental problems linked to the concentration of atmospheric particulate matter with dimensions less than 10 μm (PM_{10}) in urban settings have stimulated a variety of scientific researches. This study reports a systematic analysis of the magnetic properties of PM_{10} samples collected by six automatic stations installed for air quality monitoring through the Latium Region (Italy). We measured the low-field magnetic susceptibility of daily air filters collected during the period July 2004 to July 2005. For each station, we derived an empirical linear correlation linking magnetic susceptibility to the concentration of PM_{10} produced by local sources (i.e., in absence of significant inputs of exogenous dust). An experimental approach is suggested for estimating the percentage of nonmagnetic PM_{10} transported from natural far-sided sources (i.e., dust from North Africa and marine aerosols). Moreover, we carried out a variety of additional magnetic measurements to investigate the magnetic mineralogy of selected air filters spanning representative periods. The results indicate that the magnetic fraction of PM_{10} is composed by a mixture of low-coercivity, magnetite-like, ferrimagnetic particles with a wide spectrum of grain sizes, related to a variety of natural and anthropogenic sources. The natural component of PM_{10} has a characteristic magnetic signature that is indistinguishable from that of eolian dust. The anthropogenic PM_{10} fraction is mostly originated from circulating vehicles and is a mixture of prevailing fine superparamagnetic particles and subordinate large multidomain grains; the former are more directly related to exhaust, whereas the latter may be associated to abrasion of metallic parts.

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1. Introduction

[2] Concentration of particulate matter (PM) in the lower atmosphere is presently of particular concern in urban environments since it poses hazards and risks for the health of citizens, though the exact adverse mechanism of effect is still not fully understood [*Harrison and Yin*, 2000]. Air particles with an aerodynamic diameter of 10 μm or less (the so-called PM_{10}) pose the most serious threat to human health, since they penetrate deep into the lungs, and episodes of high concentration in the atmosphere appear to be correlated to cardiovascular and respiratory diseases and to increased cancer and mortality risks [*Morris et al.*, 1995; *Pope and Dockery*, 1999; *Wichmann and Peters*, 2000; *World Health Organization*, 2004]. As a result, long-term exposure to high PM_{10} concentration results in a substantial reduction in life expectancy [*World Health Organization*, 2004]. The problem of PM_{10} draws the vivid attention of town councils, national and international governments and, of course, the media. In 1999, the European Commission promulgated the Council Directive 99/30/EC relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, PM and lead in ambient air. Threshold values for PM_{10} were established in 50 $\mu g/m^3$ as daily average and in 40 $\mu g/m^3$ as annual average, with a forthcoming standard of 20 $\mu g/m^3$ which will be effective no sooner than 2010. In case of overcoming of these thresholds, the authorities have to enforce plans of action and counteractive measures. The directive specifies however that these thresholds are not to be applied to events defined as natural, such as resuspension of particles, long-range transport from deserts, volcanic eruptions, geothermal and seismic activities.

[3] Yearly average PM_{10} concentrations for the eight major Italian cities in 1998–1999 were estimated between 44.4 and 53.8 $\mu g/m^3$, with a population-weighted mean

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value of 52.6 $\mu g/m^3$, indicating that the societal burden of urban air pollution in Italian cities is very high [*Galassi et al.*, 2000; *World Health Organization*, 2002].

[4] Scientific interest in the understanding of the phenomenon goes along with the social interest in it, with a demand to develop innovative techniques to better characterize the sources and the spreading mechanisms of PM_{10} due to pollution and/or various natural processes. The measure and analysis of magnetic parameters appear particularly promising in this respect, since PM_{10} contains magnetic particles characterized by stable and intense magnetic properties [*Matzka and Maher*, 1999; *Maher et al.*, 1999] as a common component derived from pollution [*Weber et al.*, 2000].

[5] Concentration of magnetic PM in Munich, Germany, has been estimated as varying in the range of 0.3–0.5% by mass [*Muxworthy et al.*, 2003]. These magnetic particles, generally in the form of submicroscopic Fe-rich spherules, mostly derive from combustion processes such as industrial, domestic or vehicle emissions [*Hunt et al.*, 1984; *Flanders*, 1994, 1999] or from asphalt abrasion products and from vehicles brake systems [*Hoffmann et al.*, 1999]. Moreover, magnetic PM is associated to heavy metals like zinc, cadmium and chrome [*Georgeaud et al.*, 1997] and to mutagenic organic compounds [*Morris et al.*, 1995], all dangerous to human health.

[6] Magnetic properties, which are obtained from nondestructive, fast and low-cost analyses, provide valuable proxies to study mechanisms and processes of PM origin and dispersion in the atmosphere. In recent years, several studies employed magnetic techniques to identify atmospheric industrial and traffic pollution in cities through the magnetic analysis of PM collected on filters [*Shu et al.*, 2001; *Muxworthy et al.*, 2001, 2002, 2003; *Spassov et al.*, 2004], from street dust [*Xie et al.*, 2000; *Goddu et al.*, 2004; *Gautam et al.*, 2004; *Shilton et al.*, 2005] or from tree leaves [*Matzka and Maher*, 1999; *Hanesch et al.*, 2003; *Moreno et al.*, 2003; *Urbat et al.*, 2004]. Magnetic susceptibility, which is the parameter that can be measured more easily and rapidly, is the proxy more often used in environmental magnetic surveys.

[7] Additional studies on the PM magnetic mineral composition are carried out to identify different populations of magnetic particles and to discriminate their sources. *Spassov et al.* [2004] developed a method for quantifying anthropogenic sources based on the measurement of demagnetization curves of an anhysteretic remanent magnetization (ARM), instead of expensive chemical analyses. Their study was carried out in the urban area of Zürich (Switzerland), and additional samples were collected in a motorway tunnel. Their major conclusion was that the PM_{10} magnetic carriers at room temperature could be divided in two categories: a low-coercivity component with an ARM median destructive field (MDF) of 28 mT and a high-coercivity component (called "UP" in the following) with a MDF of 89 mT. The magnetic properties of the low-coercivity component were similar to those of natural dust, whereas the high-coercivity component correlated to the local degree of pollution, determined with chemical methods. Since the magnetization of both components increased with increasing levels of pollution, the conclusion was that the low-coercivity component is composed of a mixture of natural dust and dust resuspended by the turbulence due to circulating vehicles and other human activities [*Spassov et al.*, 2004].

[8] Former studies on the magnetic properties of widespread tree leaves in the city of Rome have shown that the source of magnetic PM in Rome is mostly due to vehicular traffic [*Moreno et al.*, 2003].

[9] This study reports a systematic examination of the magnetic properties of PM_{10} samples collected by six automatic stations operating by Regione Lazio (Italy) for continuous monitoring of air quality. Systematic measurement of magnetic susceptibility provided original data to complement the standard environmental database that constitutes the experimental ground for making decision on urban and transportation planning and for establishing environmental strategy and policy. The overall aim of this study was to set up an experimental protocol for the use of magnetic properties as reliable proxies for the identification of the natural and anthropogenic PM_{10} sources, as an attempt to define a magnetic fingerprint for various populations of fine atmospheric particles under a variety of weather and environmental conditions.

2. Air-Monitoring Stations: Location and Period of Analysis

[10] This research deals with systematic magnetic measurements carried out on daily filters collected from six selected automatic monitoring stations managed by ARPA Lazio (Agenzia Regionale Protezione Ambientale, the regional agency for environmental protection). The selected air-monitoring stations are distributed in the Latium Region (Figure 1). Two of them (Magnagrecia and Villa Ada) are within the town of Rome and represent quite different environmental conditions: Via Magnagrecia is a high-traffic road just outside the ancient Aurelian Walls, whereas Villa Ada is one of the main green park in Rome and represents the urban background. The other four stations (Viterbo, Frosinone scalo, Latina and Fontechiari) represent different cases and various scenarios of anthropogenic pollution through the Latium territory: The stations of Viterbo and Latina are within an urban environment (both stations are in provincial main towns with little industrial activity and a population of ~60,000 and 110,000 inhabitants, respectively), Frosinone scalo represents an high-traffic industrial area, and the station of Fontechiari, located in a countryside area far from pollution sources and towns, represents the rural background.

[11] The air-monitoring stations pump ~16.3 L of air per minute (that is an air volume of ~23.47 m^3/d). The daily PM_{10} is retained in a circle of 18 mm of diameter at the center of a 35 mm paper roll (glass fiber paper GF10, produced by Whatman and distributed in Italy by the Environnement Group Italia Srl, commercial code M04-370–392) which ensures retention of more than 99.9% of particulate matter with aerodynamic diameter less than 10 μm.

[12] The investigated period spans nearly one full year: Data were systematically collected from July 2004 to July 2005 (Table 1). Magnetic measurements of each daily filter have been systematically compared to the corresponding value of the recorded PM_{10} concentration, as provided from ARPA Lazio.

Figure 1. Location of the air-monitoring stations considered in this study (see text and Table 1).

[13] A companion research project, involving ARPA Lazio, the National Research Council (CNR), the University of Rome "La Sapienza" and sponsored by the Latium Region as well, was also carried out during the same period, with studies focused on the chemical analysis of the PM_{10} and on the reconstruction of atmospheric circulation to estimate natural inputs of exogenous PM (i.e., powders from North Africa). The results of these companion researches [*Regione Lazio*, 2005, 2006] will be only mentioned here when relevant to the proper understanding of the natural processes affecting the magnetic properties of PM_{10}.

3. Measurements and Laboratory Techniques

[14] The low-field magnetic susceptibility of PM_{10} filters was measured in the paleomagnetic laboratory of the Istituto Nazionale di Geofisica e Vulcanologia in Rome, using a AGICO KLY-3S kappabridge (applied magnetic field of 300 A/m, operating frequency of 0.875 kHz) calibrated to manufacturer's standard Etalon 124 to within 1 part in 1162. The clean glass fiber paper employed in this study is essentially not magnetic: The magnetic susceptibility of a 4-cm-long strip (i.e., the length of a typical filter sample including a single "circle" of daily retained PM_{10}) is not measurable on the KLY-3S kappabridge, which has a declared sensitivity of 3×10^{-8} SI.

[15] Given the relatively low magnetic signal of the studied samples, the susceptibility of each daily PM_{10} filter was computed from the average of five repeated measurements. Raw susceptibility data are normalized assuming an arbitrary volume of 10 cm^3(the nominal volume of the pick-up coil) and the resulting susceptibility k is expressed in SI units. Using daily average values of PM_{10} concentration (in $\mu g/m^3$), and taking into account the air volume pumped by the monitoring stations (23.472 m^3/d), we computed the total mass of PM_{10} collected in each daily filter (expressed in μg). We then calculated the mass-normalized susceptibility (χ, expressed in m^3/kg) multiplying the total susceptibility signal provided by the kappabridge (k_T) by the nominal volume of the pick-up coil (V_0) and dividing by the total mass of the PM_{10} sample (m):

$$\chi = (V_0/m)k_T$$

[16] The magnetic susceptibility of single daily filters collected in Fontechiari was too small to be reliably

Table 1. Statistic PM_{10} Concentration and Magnetic Susceptibility (k, χ) Values at Each Air-Monitoring Station[a]

Air-Monitoring Station	Type	Period	Number of Samples	PM_{10} Mean(sd); [max − min], $\mu g/m^3$	N[b]	k Mean(sd); [max − min], 10^{-6} SI	χ Mean(sd); [max − min], 10^{-8} m^3/kg
Viterbo	urban, small town	Jul 2004 to Jul 2005	358	31(14); [100 − 6]	28	0.55(0.30);[2.31 − 0.09]	811(298); [2048 − 267]
Villa Ada	urban background, large town	Oct 2004 to Jul 2005	277	30(14); [99 − 5]	20	0.90(0.40);[2.15 − 0.12]	1443(638); [4378 − 177]
Magnagrecia	urban, large town, high traffic	Aug 2004 to Jul 2005	334	45(16); [157 − 11]	89	2.06(0.72);[4.50 − 0.66]	2117(619); [4078 − 624]
Frosinone scalo	industrial, high traffic	Sep 2004 to Jul 2005	261	48(29); [175 − 6]	98	1.25(0.59);[4.70 − 0.14]	1304(559); [3853 − 292]
Latina	urban, small town	Sep 2004 to Jul 2005	303	29(16); [92 − 4]	15	0.50(0.26);[1.90 − 0.06]	793(498); [3077 − 82]
Fontechiari	rural background	Sep 2004 to Apr 2005	(36)[c]	24(12); [100 − 4]	12	0.46(0.21);[1.21 − 0.14][c]	20(7); [42 − 11][c]

[a] PM_{10}, k, and χ values are provided as mean(standard deviation); [max − min].
[b] N, number of days with concentration of PM_{10} higher than 50 $\mu g/m^3$ during the period 1 October 2004 to 31 July 2005.
[c] Because of the very low content of magnetic particles in the daily filters from the Fontechiari station, magnetic measurements on those filters were carried out on samples composed of groups of 7–8 each (see text).

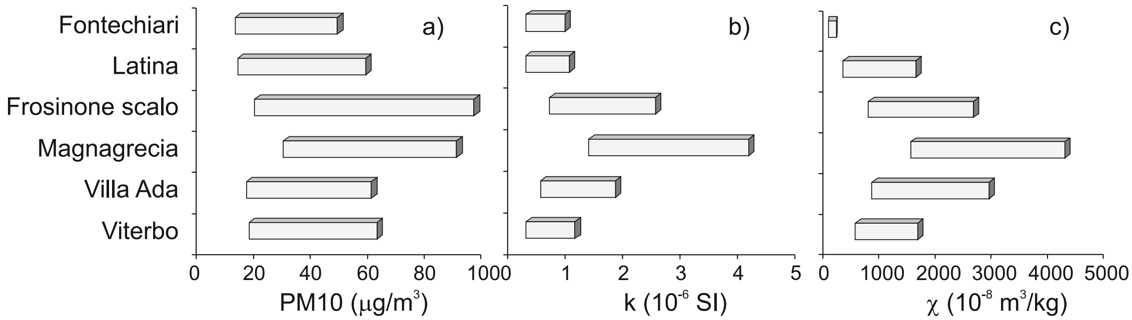

Figure 2. Range of variation for (a) the PM_{10} concentration, (b) k, and (c) χ at all air-monitoring stations. For each plot, bars are centered on the arithmetic mean values computed for the overall study period (see Table 1) and span the interval mean ±1 standard deviation. Values refer to measurements on a daily basis, except for the magnetic susceptibility (k and χ) at the Fontechiari station where they represent measurements taken on groups of 7–8 daily filters each. See text for discussion.

measured. Therefore, for each monthly batch of samples from the Fontechiari station, we measured 3 specimens composed of 7–8 daily filters each, grouping together filters characterized by high, medium and low PM_{10} concentration.

[17] A variety of additional magnetic measurements was carried out on selected groups of filters spanning representative periods, in order to investigate the details of the PM_{10} magnetic mineralogy.

[18] An ARM was imparted in an alternating field (AF) of 200 mT, with a bias field of 0.1 mT, using a pass-through superconducting rock magnetometer of the 2G Enterprises (model 755) with an in-line AF demagnetizer and DC field solenoid, within a magnetically shielded room. The ARM was successively AF demagnetized in 68 steps up to 200 mT. Both the ARM acquisition and its AF demagnetization were applied along a single axis of the samples, translating them at a constant speed of 10 cm/s through the axial coil of the AF demagnetizer. This experimental configuration results in an AF decay rate of about 67 μT/half cycle [*Sagnotti et al.*, 2003].

[19] The ARM demagnetization curves were modeled using a linear combination of appropriate functions which represent the contribution of different sources of magnetic particles to the total magnetization [*Egli*, 2003, 2004], in order to determine the coercivity distribution of the various populations of magnetic particles in a sample. For this purpose, four identically measured demagnetization curves were stacked in order to increase the signal-to-noise ratio.

[20] Hysteresis properties were measured on 4 × 4 mm square specimens, cut from the center of selected daily filters from the Magnagrecia, Villa Ada and Fontechiari stations, using a MicroMag alternating gradient magnetometer (AGM, model 2900, Princeton Measurement Corporation) with a maximum applied field of 1 T. Saturation magnetization (M_s), saturation remanent magnetization (M_{rs}) and coercive force (B_c) were determined from the hysteresis loops.

[21] Acquisition of an isothermal remanent magnetization (IRM) and subsequent DC back-field remagnetization (both in a succession of fields up to 1 T) were also carried out on the AGM on the same square specimens. Data were used to compute the coercivity of remanence (B_{cr}) and the S ratios ($S_{100} = IRM_{-100mT}/IRM_{1T}$ and $S_{300} = IRM_{-300mT}/IRM_{1T}$).

[22] First-order reversal curves (FORCs) have been obtained by cycling each selected specimen from a positive saturation field to a preparation field, with subsequent measurement in a field sweeping from the preparation field to saturation [*Pike et al.*, 1999; *Roberts et al.*, 2000]. 121 FORCs have been measured for each specimen, in steps of 2.8 mT and an averaging time of 100–500 ms (depending on the magnetic intensity of the specimen), using a 0.5 T saturating field. Correspondent FORC diagrams have been calculated using the FORCobello software written by M. Winklhofer (available at http://venus.geophysik.uni-muenchen.de/~michael/forcnew/). FORC diagrams are contour plots describing the coercivity distribution of magnetic particles and their interaction field strengths [*Pike et al.*, 1999; *Roberts et al.*, 2000].

[23] Finally, samples from Villa Ada (8–11 October 2004) and Fontechiari (10–18 October 2004) were subjected to low-temperature experiments at the Institute for Rock Magnetism (Minnesota). The in-phase (χ') and out-of-phase (χ'') susceptibility, as well as thermal demagnetization between 4 K and 320 K of zero-field-cooled and field-cooled remanences acquired in a 2.5 T field, were measured using a magnetic property measurement system (MPMS, Quantum Design).

4. Results

4.1. Magnetic Susceptibility: Intensity

[24] Average values of k at the different stations range from 0.5 to 2.1 (10^{-6} SI). A basic statistical analysis of the PM_{10} concentration and magnetic susceptibility data from the various stations (Table 1 and Figure 2) shows the following:

[25] 1. PM_{10} concentrations are highest in the stations located along high-traffic roads (Magnagrecia) and in industrial areas (Frosinone scalo). Similar and consistently lower PM_{10} concentrations characterize urban environments of small towns (Viterbo, Latina), the urban background of Rome (Villa Ada), and the regional rural background (Fontechiari).

[26] 2. The highest values of k were measured in Magnagrecia and Frosinone scalo, and the lowest in Fontechiari (rural background). In Fontechiari, 7–8 days are generally necessary to increase k to values comparable with the daily

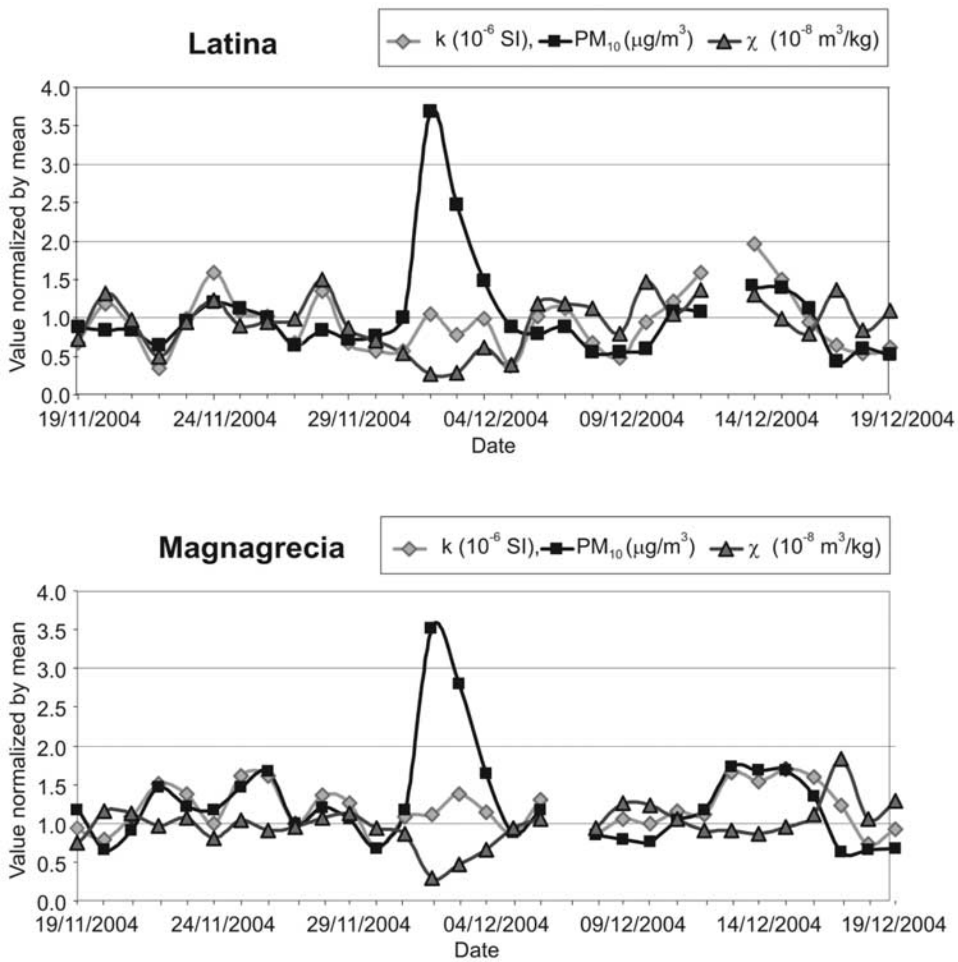

Figure 3. Representative plots of daily variation of the low-field magnetic susceptibility normalized by unit volume (k, in 10^{-6} SI) and by unit mass (χ, in 10^{-8} m^3/kg), and of the mean daily concentration of PM_{10} (in μg/m^3). The plots refer to the PM_{10} filters sampled at the Latina and Magnagrecia stations during the period 19 November to 19 December 2004. See text for discussion.

k values measured in the urban environment of small towns (Viterbo, Latina).

[27] 3. The mass-normalized magnetic susceptibility χ is highest in Rome (even in the main urban park of Villa Ada) and at Frosinone scalo. The χ values observed in Fontechiari are 2 orders of magnitude lower than those measured at Magnagrecia.

[28] 4. The variability of PM_{10}, k, and χ, computed as standard deviation, is typically ~50% of the arithmetic mean values.

4.2. Magnetic Susceptibility Versus PM_{10} Concentration: Temporal Trends

[29] The comparison of the daily variation of the magnetic susceptibility values and the PM_{10} concentrations at each station indicates that the two parameters appear often, but not always, correlated. A typical example of monthly k and PM_{10} variation is shown in Figure 3, for the Latina and Magnagrecia stations (19 November to 19 December 2004). In general, we distinguish three different cases (as exemplified in Figure 3):

[30] 1. In case A, PM_{10} concentration vary as k (χ is almost constant). This indicates that the magnetic composition of PM_{10} is constant over time. This case represents most of the investigated period.

[31] 2. In case B, the PM_{10} concentration increases more than (or decreases less than) k. These are episodes of χ minima associated to a relative increase of the nonmagnetic fraction of PM_{10}, suggesting an input of exogenous dust. The main episode occurred on 2–4 December 2004 (see Figure 3).

[32] 3. In case C, PM_{10} concentration decreases more (or increases less) than k (i.e., 16–18 December 2004 in Figure 3). These episodes of χ maxima occurred mostly during periods of low PM_{10} concentration and indicates a preferential removal of nonmagnetic PM (i.e., a relative increase of the magnetic fraction of PM_{10}) during periods of clean atmospheric conditions. This case generally corresponds to the main rain episodes.

[33] To empirically assess the relationship between the PM_{10} concentration and the magnetic susceptibility k under typical meteorological conditions (i.e., case A), we selected, for each month and at all stations, the days characterized by a relatively constant χ. As a selecting criterion, we chose daily values of χ comprised within ±20% of the monthly

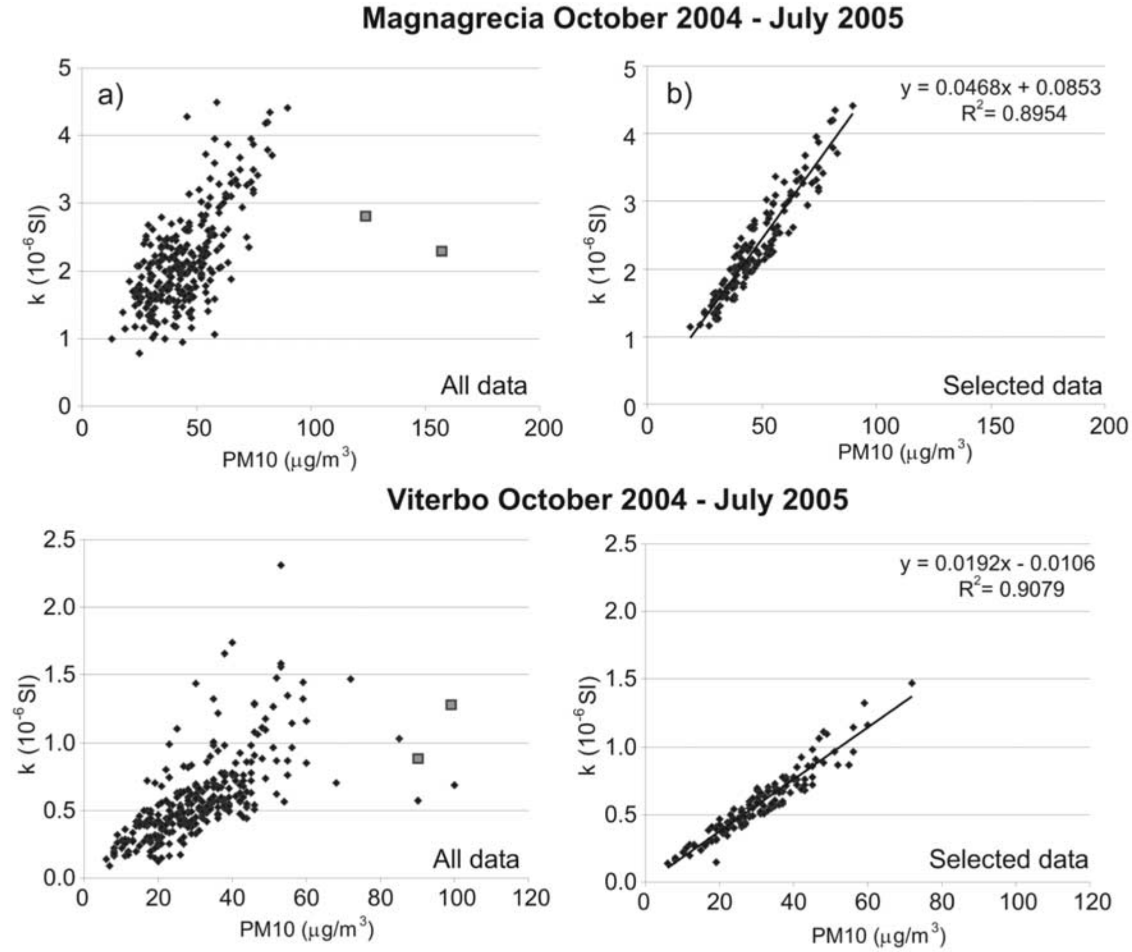

Figure 4. Concentration of PM_{10} versus the low-field magnetic susceptibility (k) at the Magnagrecia and Viterbo stations. (a) All available data. Squares indicate the data for days 2–3 December 2004. (b) Data that were used to derive an empirical linear correlation of k versus PM_{10} concentration. At all stations such correlation was estimated for selected days with χ comprised within ±20% of the arithmetic mean value.

average. The selected days were then used to estimate an empirical linear correlation between k and PM_{10} concentration (expressed by $k = a\ PM_{10} + b$), with k in 10^{-6} SI and PM_{10} in $\mu g/m^3$ (Figure 4). The coefficients a and b depend on local conditions and differ from station to station (Table 2). The proportionality constant a, computed for monthly intervals, is relatively constant at each station for the whole period of observation and is highest for the two stations in Rome (Magnagrecia, Villa Ada). The steadiness of the correlation coefficients is particularly evident for the two stations with the highest magnetic susceptibility values and PM_{10} concentrations (Magnagrecia and Frosinone scalo) (Table 3). A remarkable change of the coefficient a was observed for the Latina station, around late February to early March 2005 (Figure 5). We assumed that some change occurred in the environmental conditions around the station and preferred to consider the two periods October 2004 to February 2005 and March–July 2005 as distinct (Table 2).

[34] We used the empirical linear correlation ($k = a\,PM_{10} + b$) to compute the PM_{10} concentration correlated to the

Table 2. Empirical Linear Correlations of k Versus PM_{10} Concentration[a]

Station	Period	N (N_{tot})	Slope a	Intercept b	R^2
Viterbo	Oct 2004 to Jul 2005	125 (301)	0.019	−0.011	0.91
Villa Ada	Oct 2004 to Jul 2005	75 (183)	0.031	0.135	0.88
Magnagrecia	Oct 2004 to Jul 2005	149 (273)	0.047	0.085	0.90
Frosinone scalo	Nov 2004 to Jul 2005	87 (216)	0.022	0.228	0.94
Latina	Oct 2004 to Feb 2005	46 (139)	0.021	0.086	0.92
	Mar to July 2005	62 (129)	0.010	−0.005	0.89

[a]The correlation $k = a\ PM_{10} + b$, with k in 10^{-6} SI and PM_{10} in $\mu g/m^3$. N, number of days selected to compute the linear correlations; N_{tot}, total number of days.

Table 3. Monthly Empirical Linear Correlations of *k* Versus PM_{10} Concentration for the Magnagrecia Station[a]

Period	N (N_{tot})	Slope	Intercept	R^2
Oct 2004	16 (28)	0.043	0.180	0.76
Nov 2004	18 (29)	0.044	0.459	0.87
Dec 2004	11 (18)	0.042	0.283	0.90
Jan 2005	25 (31)	0.045	0.271	0.92
Feb 2005	17 (27)	0.045	0.673	0.89
Mar 2005	16 (30)	0.035	0.730	0.65
Apr–May 2005	28 (65)	0.045	0.138	0.64
Jun 2005	13 (28)	0.038	0.237	0.78
Jul 2005	18 (26)	0.032	0.332	0.79
Oct 4 to Jul 2005	149 (273)	0.047	0.085	0.90

[a]The correlation $k = a\ PM_{10} + b$, with k in 10^{-6} SI and PM_{10} in $\mu g/m^3$. N, number of days selected to compute the linear correlations; N_{tot}, total number of days.

magnetic susceptibility *k* values on a daily basis. We further define the "residual nonmagnetic" PM_{10} concentration (PM_{10NM}) as the amount of PM_{10} that is not linearly correlated to the magnetic susceptibility *k*. This parameter provides an original estimate of the exogenous nonmagnetic fine atmospheric particles inputs from natural sources.

[35] As an example, Figure 6 shows the daily variation of the measured PM_{10} concentration and the estimated PM_{10NM} for four stations during the period October–December 2004. The pie diagrams in Figure 7 show the chemical composition of PM_{10} for episodes of exogenous inputs of marine aerosol (19 November 2004), terrigenous dust from Sahara (1–4 December 2004) and events of secondary pollution (6 November 2004) [*Regione Lazio*, 2005]. We point out the similarity of the estimated PM_{10NM} concentration values at the various stations and the consistency of these values with those computed from chemical analyses (Figures 6 and 7).

4.3. ARM and Coercivity Distribution

[36] The ARM magnetic moment of the samples ranges from 4×10^{-10} A m^2/d (Fontechiari) to 3×10^{-9} A m^2/d (Magnagrecia, Frosinone). The ARM of a strip of clean glass fiber paper with a length of 4 cm (equivalent to the length of a typical daily PM_{10} sample) is of the order of 1.5×10^{-10} A m^2 and is completely demagnetized by 50 mT AF. An example of ARM demagnetization curve for a PM_{10} sample is shown in Figure 8a. Coercivity distributions were calculated, according to *Egli* [2003], for samples from the Fontechiari, Latina, Frosinone scalo and Magnagrecia stations (Figure 8b). A component analysis was performed, according to *Egli* [2004], on selected coercivity distributions obtained from the ARM demagnetization measurements, and, in one case (Magnagrecia) from the DC back-field remagnetization of a forward saturation IRM (Figure 9). Three distinct magnetic components have been identified from the ARM measurements (Figure 9a), and an additional high-coercivity component results from the IRM DC remagnetization curve (Figure 9b). Properties of the magnetic components are consistent in all the samples analyzed. Results obtained from ARM and the IRM DC remagnetization curves for Magnagrecia are also consistent: The small differences between the MDF of the corresponding components can be attributed to the different type of magnetization investigated (ARM versus IRM [see *Egli and Lowrie*, 2002]) and the different demagnetization techniques [*Cisowski*, 1981].

[37] The results of component analysis performed on ARM demagnetization curves, for various sites and periods, are reported in Table 4. Measurements have been fitted using three components (indicated as ARM1, ARM2 and ARM3 in Figure 9a and in Table 4), since each of these components is individually recognizable in several coercivity distributions calculated from the measurements. The

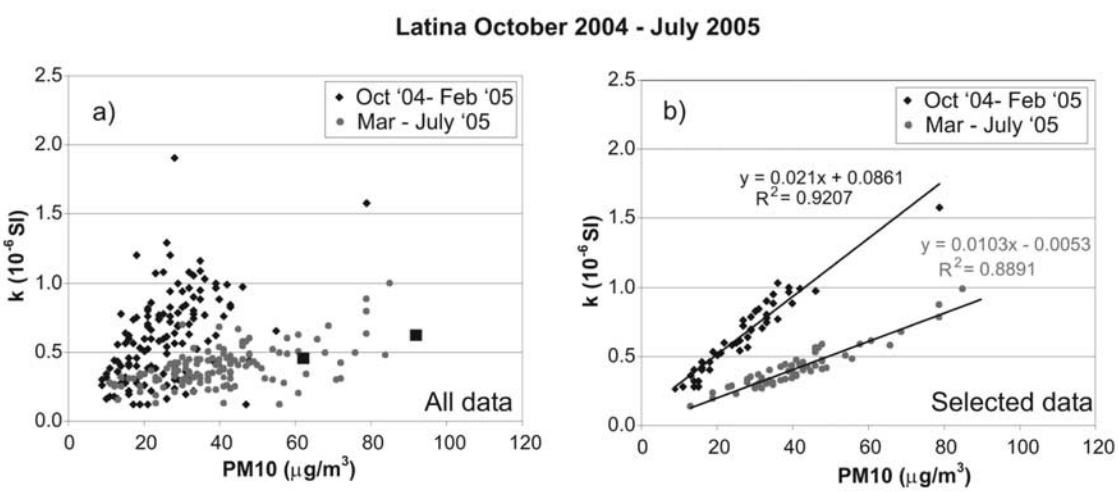

Figure 5. Concentration of PM_{10} versus the low-field magnetic susceptibility (*k*) at the Latina station. (a) All available data. Squares indicate the data for days 2–3 December 2004. (b) Data that were used to derive an empirical linear correlation of *k* versus PM_{10} concentration, selecting days with χ comprised within ±20% of the arithmetic mean value. At this station a clear change in the relationship between PM_{10} and *k* was observed around late February to early March 2005. A linear correlation between the two parameters was therefore computed for two distinct periods (Figure 5b): October 2004 to February 2005 and March–July 2005. This change has to be linked to a permanent variation of the local environmental conditions.

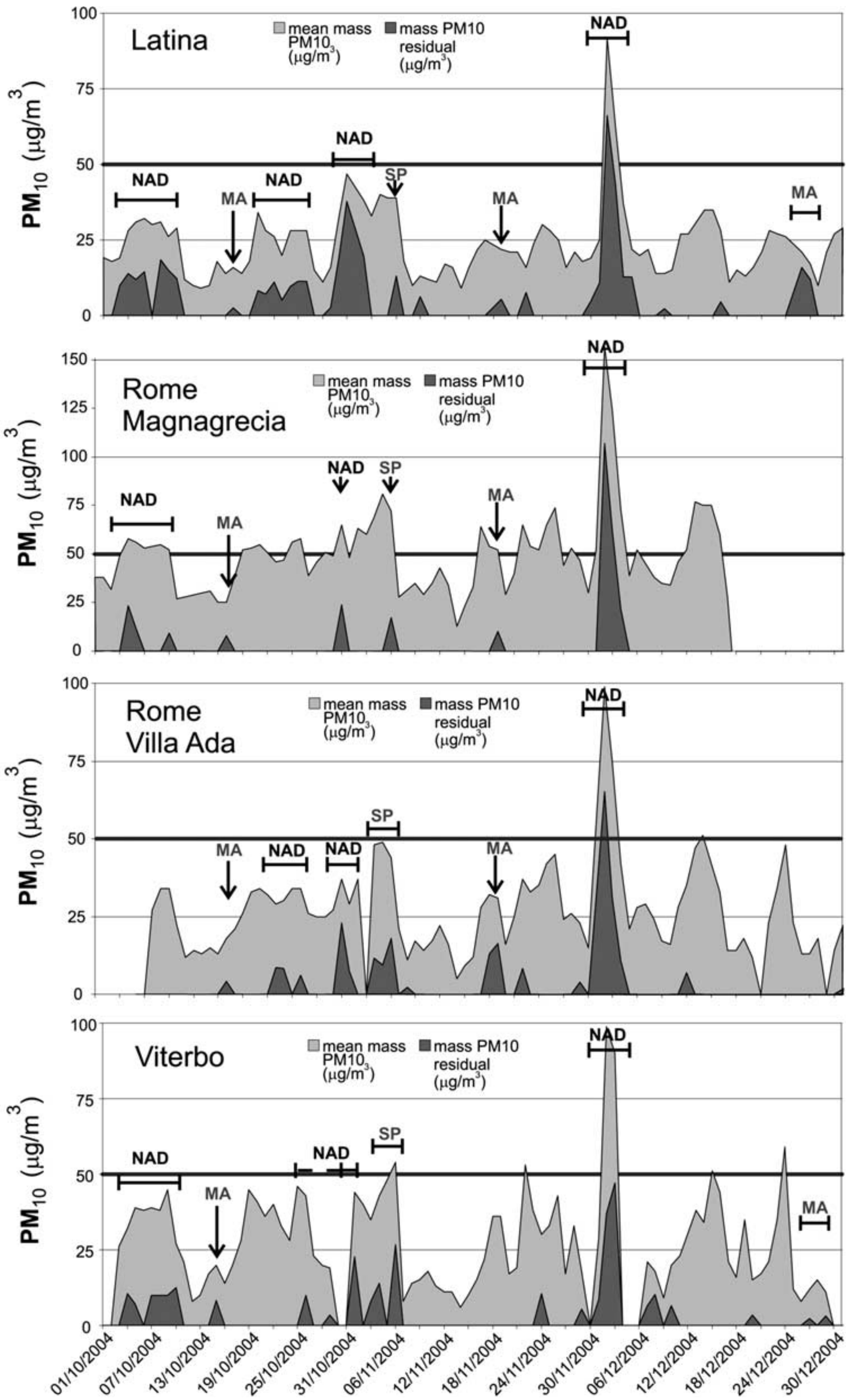

Figure 6. Diagrams for the period October–December 2004 and the daily variation of the measured PM_{10} concentration and the estimated "residual not magnetic" PM_{10} concentration for four stations. We point out the similarity of the estimates for residual not magnetic PM_{10} at all stations during the same events for which a natural contribution is known [*Regione Lazio*, 2005]. MA, marine aerosol; NAD, North African dust; SP, secondary pollution.

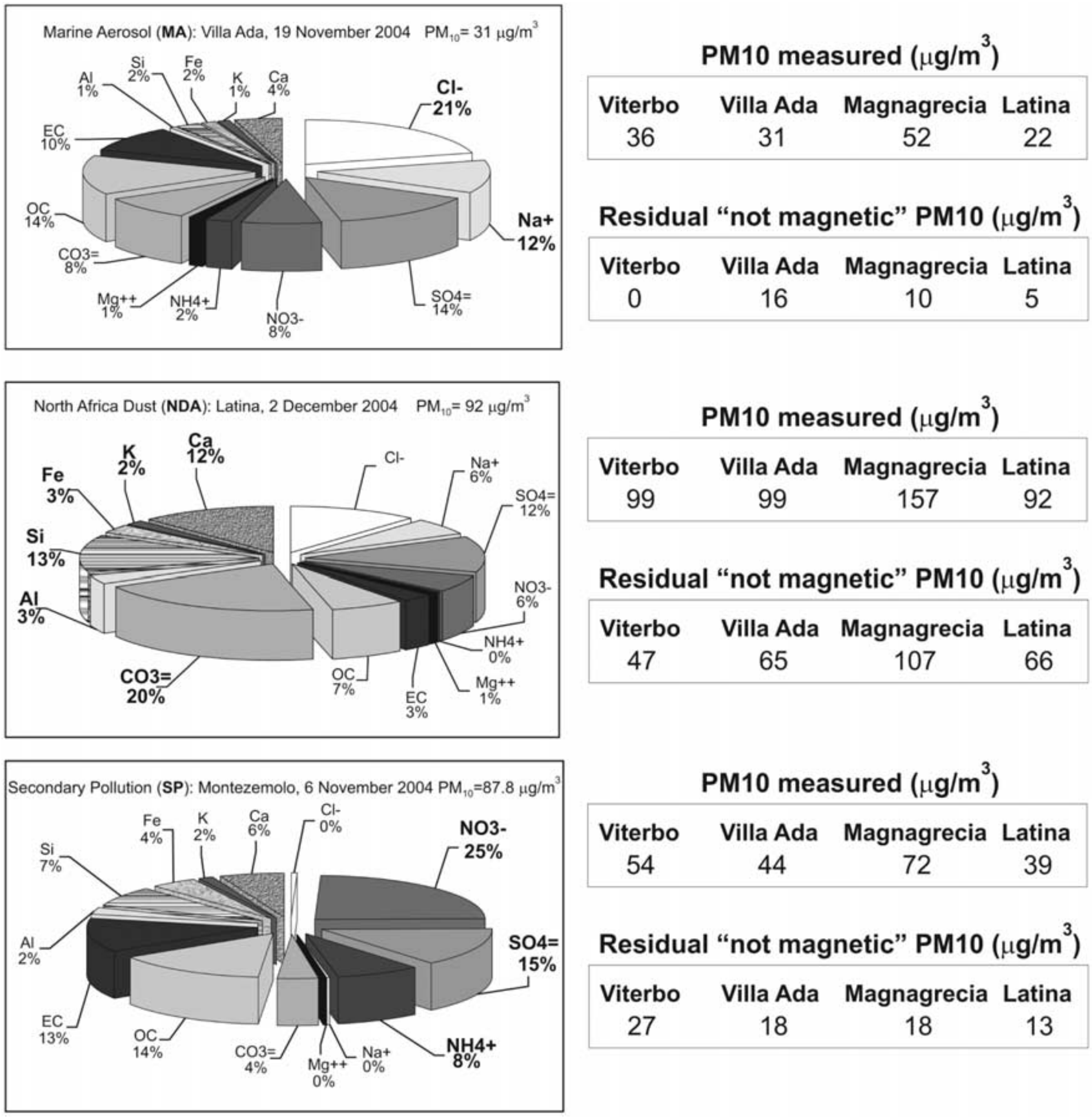

Figure 7. Pie diagrams show the chemical composition of PM_{10} [*Regione Lazio*, 2005] for the major of events of "not magnetic" PM_{10} inputs during the same period shown in Figure 6. In each pie diagram, values in bold indicate the main elements that may be linked to the outlined events.

intrinsic properties of the components, summarized by the MDF and the dispersion parameter (DP), are very consistent through the entire set of samples. The ARM3 component (MDF $\approx$ 100 mT, DP $\approx$ 0.2) has the same properties of the "UP" component attributed to motor vehicle emissions by *Spassov et al.* [2004]. All stations, but Fontechiari, show a similar magnetic composition in terms of the three ARM components, and no particular dependence of MDF and DP on the site or the sampling period was found. Even during periods of maximum North African dust inputs (e.g., 1–4 December 2004, see also Figure 3) the same components with identical coercivity distributions are observed.

[38] Instead, the magnetic composition of Fontechiari is highly variable in the two analyzed periods. The data for the 10–18 October 2004 interval characterized by a low PM_{10} concentration, could be fitted by a single component (ARM2) only, whereas those for the 5–14 January 2005 period are better fitted by the same three ARM components recognized at the other stations.

4.4. Hysteresis Properties and FORC Diagrams

[39] Hysteresis data for selected samples are reported in Table 5 and shown in Figure 10. The hysteresis properties indicate that the magnetic grains are composed of low-coercivity magnetite-like minerals: Saturation is reached in fields of 300 mT and the coercivity of remanence is typically within the 35–40 mT range. The hysteresis ratios (M_{rs}/M_s and B_{cr}/B_c), for the selected Magnagrecia and Villa Ada filters, indicate a fairly homogeneous grain size distribution, with data plotting in a restricted region of the Day plot [*Day et al.*, 1977; *Dunlop*, 2002a, 2002b] in between the mixing trends for single-domain (SD)/multidomain (MD) and SD/superparamagnetic (SP) magnetite grains (Figure 10).

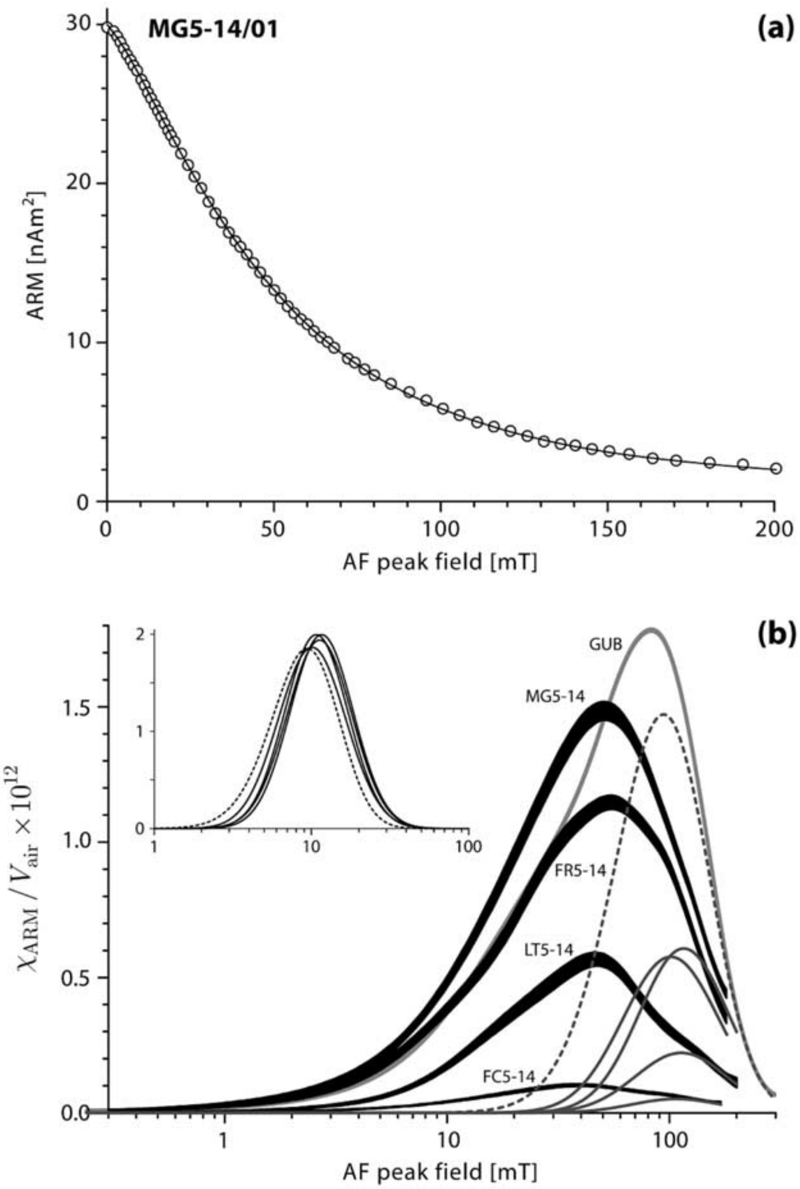

Figure 8. (a) Example of ARM demagnetization curve used for the component analysis (Magnagrecia, 5–14 January 2005). The curve is the average of four independent AF demagnetization measurements of an ARM acquired in a 0.1 mT DC field superimposed on a 200 mT AF. The solid line is the fitted demagnetization curve obtained with the program CODICA [*Egli*, 2003]. (b) Coercivity distributions calculated from ARM demagnetization curves, such as in Figure 8a, for a selection of samples collected during 5–14 January 2005 (solid curves). The thickness of the curves corresponds to the measurement error estimated automatically by CODICA. Sample GUB, collected in the center of a motorway tunnel [*Spassov et al.*, 2004] is shown for comparison. Solid thin lines correspond to the component UP (see text for detail), as obtained from a component analysis after *Egli* [2003]. The dashed thin line is the same component in GUB [*Spassov et al.*, 2004]. The insert shows the coercivity distributions of component UP normalized to a unit magnetization. The unitless coercivity distribution in Figure 8b is obtained by normalizing the measured ARM moment (A m^2) by the DC bias magnetic field (79.57 A/m) and by the volume of filtered air (23.472 m^3/d, multiplied by the number of sampled days).

[40] Remarkably different is the datum from the Fontechiari station that falls within the field for pseudosingle-domain (PSD) grains, in the region of the Day plot typical for the natural eolian deposits of northern midlatitudes (Figure 10). In the Magnagrecia station, a subtle difference in the B_{cr}/B_c ratio distinguishes the days representing high-pollution periods (lower B_{cr}/B_c) from those characterized by significant inputs of exogenous dust (higher B_{cr}/B_c) (see Table 5).

[41] The FORC diagrams clearly show a mixture of two magnetic contributions (Figure 11). The first is represented

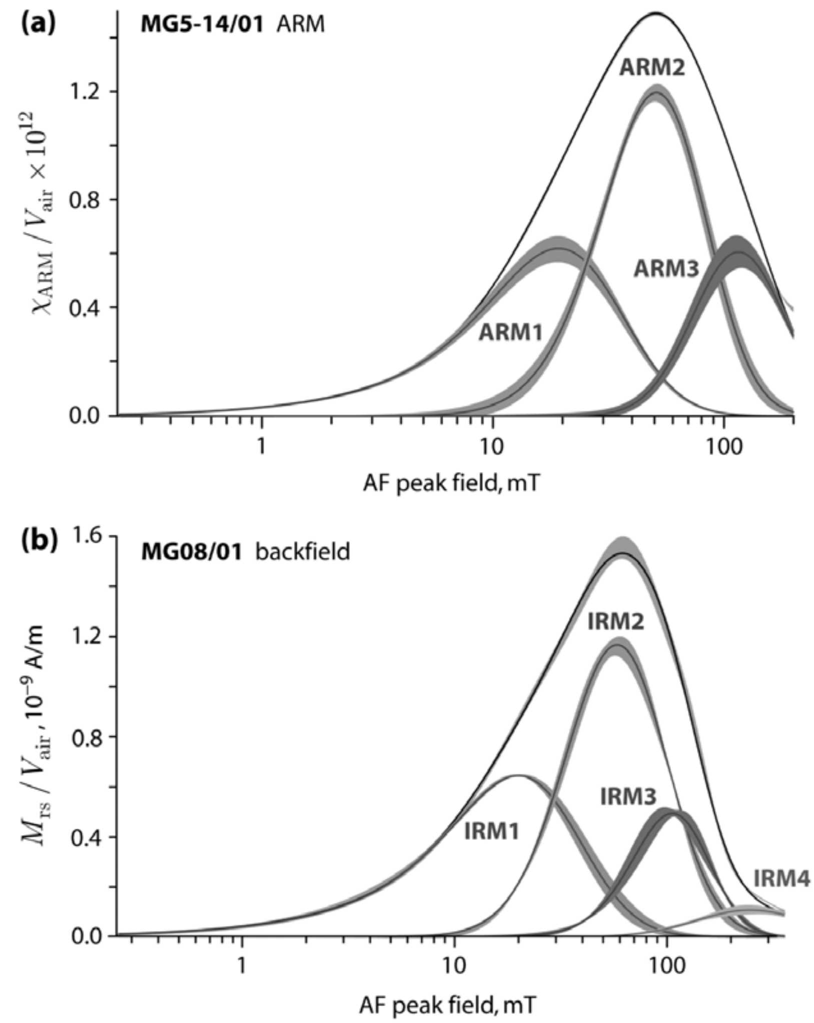

Figure 9. Two examples of component analysis after *Egli* [2003] for (a) the AF demagnetization of ARM for sample MG5-14/01 (Magnagrecia, 5–14 January 2005) and (b) the DC back-field remagnetization of IRM for sample MG08/01 (Magnagrecia, 8 January 2005). The thickness of the lines corresponds to the error estimated by the used software [*Egli*, 2004]. Notice the similarity of the results in Figures 9a and 9b, which proves the consistency of the component analysis. An additional component in Figure 9b was used to fit the magnetization curve at higher fields. The components are labeled according to Table 4.

by a coercivity distribution elongated along the B_c axis, while the second is concentrated along the B_b axis. The diagrams show a maximum peak near the origin of the plot which indicates a prevailing SP behavior. This configuration is similar to that obtained from samples containing a substantial amount of viscous magnetization, such as the Yucca Mountain Tuff CS914 and the Cretaceous red sediments [see *Roberts et al.*, 2000, Figure 6]. This result is confirmed by low-temperature measurements (see next paragraph).

[42] We point out that, in agreement with the general hysteresis results, FORC diagrams are remarkably similar for periods characterized by main inputs of north African dust (Figure 11a, 3 December 2004) and periods characterized by a stagnant atmosphere and high pollution (Figure 11b, 12 January 2005).

4.5. Low-Temperature Measurements

[43] Low-temperature measurements have been performed for selected specimens from the Fontechiari and Villa Ada stations and for the PM_{10} sample GUB collected in a motorway tunnel by *Spassov et al.* [2004]. Thermal demagnetization curves of an IRM, imparted in 2.5 T at 4 K, are characterized by a strong decay of the magnetization, with a M_r(4K)/M_r(320K) of 8.3, 4.4, and 2.4 for GUB, Villa Ada, and Fontechiari, respectively (Figure 12a). These ratios suggest differences in the magnetic mineral populations of the three samples that can be explained by different relative concentration of SP grains. A positive correlation between the concentration of SP grains and the degree of pollution is also suggested by the frequency dependence of susceptibility χ_{fd} (Figure 12b). The temperature dependence of χ_{fd} shows clear differences among the three samples. In Fontechiari, $\chi_{fd}(T)$ is a monotonically increasing function of

Table 4. Results of the Component Analysis of Detailed ARM Measurements for Selected Stations and Periods[a]

Station/Date	ARM1, %	ARM2, %	ARM3, %	MDF1, mT	MDF2, mT	MDF3, mT	DP1	DP2	DP3	Notes
Magnagrecia	*34.5 (0.8)*	*46.5 (0.5)*	*19.0 (0.2)*	*14.8 (0.8)*	*47.1 (3)*	*113 (6)*	*0.379 (0.06)*	*0.222 (0.03)*	*0.182 (0.02)*	Rome (high traffic)
1–4 Dec 2004	34.0	46.9	19.1	14.3	45.3	109	0.337	0.204	0.164	North African dust
5–14 Jan 2005	35.1	46.2	18.8	15.4	48.9	118	0.421	0.240	0.199	
Villa Ada	*40.8 (–)*	*39.7 (–)*	*19.6 (–)*	*14.3 (–)*	*45.7 (–)*	*103 (–)*	*0.403 (–)*	*0.229 (–)*	*0.222 (–)*	Rome (park)
5–14 Jan 2005	40.8	39.7	19.6	14.3	45.7	103	0.403	0.229	0.222	
Latina	*30.9 (3)*	*49.6 (6)*	*19.5 (6)*	*15.5 (0.5)*	*43.7 (3)*	*114 (8)*	*0.357 (0.01)*	*0.228 (0.01)*	*0.181 (0.04)*	urban (small town)
5–14 Jan 2005	28.6	47.1	24.3	15.2	40.8	105	0.353	0.239	0.217	
23–27 Jan 2005	33.8	44.8	21.4	16.1	45.8	120	0.370	0.231	0.186	
15–18 Mar 2005	30.3	56.8	12.9	15.2	44.4	117	0.349	0.214	0.140	
Viterbo	*26.0 (1)*	*58.1 (0.1)*	*15.8 (1)*	*12.7 (0.1)*	*40.8 (2)*	*120 (1)*	*0.380 (0.05)*	*0.247 (0.01)*	*0.170 (0.02)*	urban (small town)
5–14 Jan 2005	26.9	58.2	14.9	12.8	42.1	120	0.416	0.254	0.157	
23–27 Jan 2005	25.1	58.1	16.8	12.7	39.6	121	0.344	0.240	0.182	
Frosinone Scalo	*33.2 (2)*	*44.4 (3)*	*22.5 (3)*	*14.6 (1)*	*46.5 (1)*	*110 (9)*	*0.405 (0.02)*	*0.233 (0.01)*	*0.201 (0.02)*	industrial (high traffic)
5–14 Jan 2005	31.0	43.5	25.5	13.2	45.5	100	0.406	0.243	0.214	
23–27 Jan 2005	34.7	42.2	23.1	15.1	47.3	118	0.424	0.224	0.209	
15–18 Mar 2005	33.8	47.4	18.8	15.5	46.8	113	0.386	0.232	0.180	
Fontechiari										rural background
10–18 Oct 2004	-	100	-	-	42.8	-	-	0.32	-	
5–14 Jan 2005	31.3	45.7	23.1	11.7	37.8	107	0.449	0.240	0.200	
All stations										
5–14 Jan 2005	*32.3 (5)*	*46.7 (6)*	*21.0 (4)*	*13.8 (1)*	*43.5 (4)*	*109 (8)*	*0.408 (0.03)*	*0.241 (0.01)*	*0.201 (0.02)*	period of high pollution
23–27 Jan 2005	*31.2 (5)*	*48.4 (9)*	*20.4 (3)*	*14.6 (2)*	*44.2 (4)*	*120 (2)*	*0.379 (0.04)*	*0.230 (0.01)*	*0.192 (0.01)*	period of low PM_{10}

[a]ARM1, ARM2, ARM3 are the relative contributions of the three magnetic components (see text) to the total ARM. MDF and DP are the median destructive fields and dispersion parameters, respectively. Italic numbers are averages of the same station or the same period. Numbers in parentheses are standard deviations.

Table 5. Hysteresis Properties of Selected Filters[a]

Station/Date	M_s, 10^{-9} A m^2	M_{rs}, 10^{-9} A m^2	B_c, mT	B_{cr}, mT	M_{rs}/M_s	B_{cr}/B_c	S_{-100}	S_{-300}	χ_{hys}/M_s, 10^{-6} m/A	Notes
Magnagrecia										
31 Oct 2004	85	7	6.5	37.5	0.08	5.80	−0.73	−1.00	15.1	input of North African dust (peak)
19 Nov 2004	115	9	6.5	39.2	0.08	6.02	−0.72	−1.00	14.7	input of marine aerosol
3 Dec 2004	155	12	6.2	40.3	0.08	6.46	−0.69	−1.00	15.8	major input of North African dust (peak)
7 Jan 2005	107	8	7.4	39.7	0.08	5.36	−0.69	−1.00	12.5	period of high pollution (5–14 Jan 2005)
12 Jan 2005	239	19	7.5	40.4	0.08	5.36	−0.69	−1.00	12.4	period of high pollution (5–14 Jan 2005)
25 Jan 2005	238	19	6.5	38.2	0.08	5.85	−0.70	−1.00	14.9	period of low k and PM_{10}, χ maximum (23–27 Jan 2005)
18 Mar 2005	188	17	7.5	38.2	0.09	5.06	−0.76	−1.00	14.4	period of high pollution (13–18 Mar 2005)
21 Jul 2005	58	5	7.2	37.6	0.09	5.26	−0.73	−1.00	15.2	minor input of North African dust
Fontechiari										
10–18 Oct 2004	50	12	14.3	42.2	0.24	2.96	−0.78	−0.99	9.6	period of low k and PM_{10}; χ maximum
Villa Ada										
31 Oct 2004	70	6	10.2	42.1	0.09	4.12	−0.65	−0.99	10.7	input of North African dust (peak)
19 Nov 2004	85	7	7.3	43.6	0.09	5.94	−0.70	−0.99	14.0	input of marine aerosol
3 Dec 2004	91	10	8.9	43.7	0.10	4.91	−0.65	−1.00	14.2	major input of North African dust (peak)
7 Jan 2005	57	7	9.8	41.8	0.12	4.26	−0.61	−1.00	11.5	period of high pollution (5–14 Jan 2005)
12 Jan 2005	108	12	9.1	42.7	0.11	4.71	−0.69	−1.00		period of high pollution (5–14 Jan 2005)
25 Jan 2005	11	1	8.5	40.0	0.11	4.69	−0.61	−1.00		period of low k and PM_{10}, χ maximum (23–27 Jan 2005)
18 Mar 2005	165	11	7.5	42.5	0.07	5.64	−0.67	−−1.00		period of high pollution (13–18 Mar 2005)
Gubrist tunnel										
Spassov et al. [2004]	197	18	9.6	52.2	0.094	5.463	0.793	0.992	11.1	center of a highway tunnel

[a]Moments of samples consisting of filters collected during several days are normalized to the number of days. Moment of a sample collected in the Gubrist tunnel is normalized to a volume of 23.472 m^3 of filtered air.

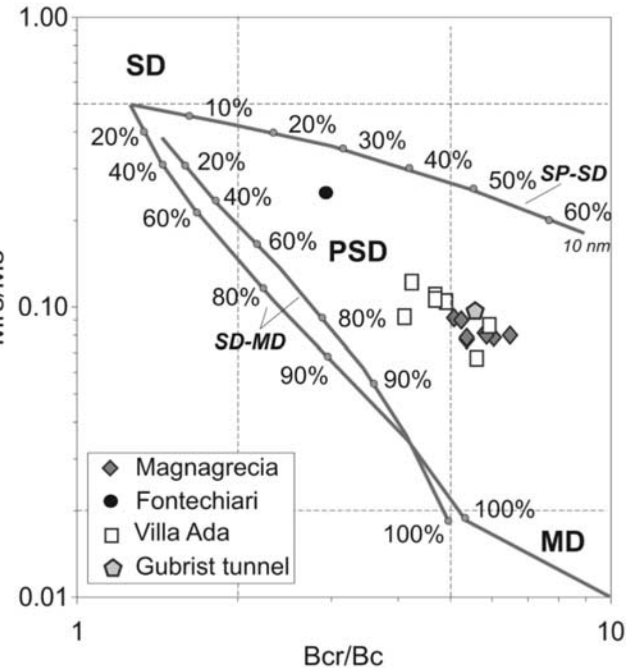

Figure 10. Hysteresis ratios (M_{rs}/M_s and B_{cr}/B_c) for Magnagrecia, Villa Ada, and Fontechiari filters (see Table 5), plotted according to *Day et al.* [1977]. The data fall in between the theoretical curves, indicated by the solid lines, calculated for mixtures of single domain (SD) and multidomain (MD) and of SD and superparamagnetic (SP) (titano)magnetite grains [*Dunlop*, 2002a]. Fields for SD, pseudosingle-domain (PSD), and MD grains are also shown after *Dunlop* [2002a]. The datum for the Fontechiari sample is clearly distinct from those from the two air-monitoring stations in Rome. See text for discussion.

the temperature, as commonly observed in soils [e.g., *Liu et al.*, 2004a], whereas Villa Ada and GUB show a distinctive broad maximum of $\chi_{fd}(T)$ centered on $T \approx 160$ K that presumably corresponds to the mean blocking temperature of anthropogenic SP particles.

5. Discussion

[44] The overall magnetic susceptibility data indicate that the concentration of magnetic particles in PM_{10} is mostly related to fractions originated from anthropogenic pollution, with a distinct higher content of magnetic particles in the total PM_{10} of the largest urban area (Rome; stations of Magnagrecia and Villa Ada) and of the industrial setting of Frosinone scalo. The PM_{10} far from pollution sources (Fontechiari) is very poorly magnetic.

[45] The empirical linear correlations computed at each station (but Fontechiari) allowed an original estimate of the inputs of exogenous PM_{10}, as indicated by the amount of PM_{10NM}.

[46] Natural exogenous PM in the Latium Region has been recognized to be often composed of marine aerosols (i.e., salt) and dust from the arid region of North Africa [*Regione Lazio*, 2005, 2006). Saharan dust mainly consists of a mixture of paramagnetic and diamagnetic minerals such as quartz, feldspars, and clays, sometimes associated with calcite and/or palygorskite [*Caquineau et al.*, 2002]. The order of abundance of the different minerals is estimated as follows: illite > quartz > smectite > palygorskite > kaolinite > calcite > dolomite > feldspars [*Avila et al.*, 1997].

[47] The whole set of measurements carried out in this study shows that the magnetic properties of PM_{10} are not influenced by the presence of high concentration of North African dust, whose magnetic susceptibility is negligible. The marine aerosols are also magnetically negligible, since sea salt is almost diamagnetic.

[48] The analysis of the PM_{10NM} concentration through the study period indicates that events of significant PM_{10NM} inputs have been relatively frequent in the trimester October–December 2004, with an estimated PM_{10NM} concentration higher than 5 $\mu g/m^3$ in more than 2 stations for 29 days (32% of the period). Of particular relevance is the episode of powder input from North Africa of 2–3 December 2004, with an estimated PM_{10NM} concentration between 50 and 100 $\mu g/m^3$ at all stations (Figures 3–5). Remarkably, a total PM_{10} concentration of 100 $\mu g/m^3$ was recorded on 2 December 2004 at the Fontechiari station.

[49] In the first half of 2005, events of significant PM_{10NM} inputs have been less frequent; days with an estimated PM_{10NM} concentration higher than 5 $\mu g/m^3$ in more than 2 stations were only 12 for the trimester January–March 2005 (13% of the period) and 19 for the interval April–July 2005 (16% of the period). These episodes, however, never reached the climax of 2–3 December 2004 and are characterized by a PM_{10NM} concentration estimated in the range of 5–15 $\mu g/m^3$, with maximum peaks of $\sim$40 $\mu g/m^3$ (26–27 March, 25 April, 6–8 July).

[50] The concentration of the magnetic mineral fraction in the PM_{10} throughout the Lazio Region appears as a reliable proxy of local air pollution. The whole magnetic data set indicates that the composition and the grain size distribution of the PM_{10} magnetic mineral fraction is rather uniform at the different stations, but Fontechiari where the influence of pollution sources appears very low. The comparison between the magnetic data obtained at Fontechiari and those obtained at all the other stations indicates that the magnetic properties of natural and anthropogenic contributions are actually different. Hysteresis and low-temperature properties of the Fontechiari filters spanning the period 10–18 October 2004 are not distinguishable from those of typical eolian deposits at midlatitude, both at room temperature (Day diagram [*Liu et al.*, 2004b], component analysis [*Spassov et al.*, 2003; *Egli*, 2004]) and at low temperature (susceptibility [*Liu et al.*, 2004a]). On the other hand, all magnetic data are consistent with the prevalence of a low-coercivity, magnetite-like, ferrimagnetic phase in the PM_{10} originated from pollution sources, with a prevalence of ultrafine SP particles in a mixture with large MD grains.

[51] Low-temperature experiments indicate that only <15% of the magnetization carried by magnetic minerals contained in anthropogenic PM_{10} emissions is blocked at room temperature (as evident from the data referred to GUB in Figure 12). This result suggests that the most precise magnetic characterization parameters for the quantification of pollution in PM_{10} rely on measurements that are sensitive to the widest range of grain sizes, such as M_s and remanence magnetizations at 4K.

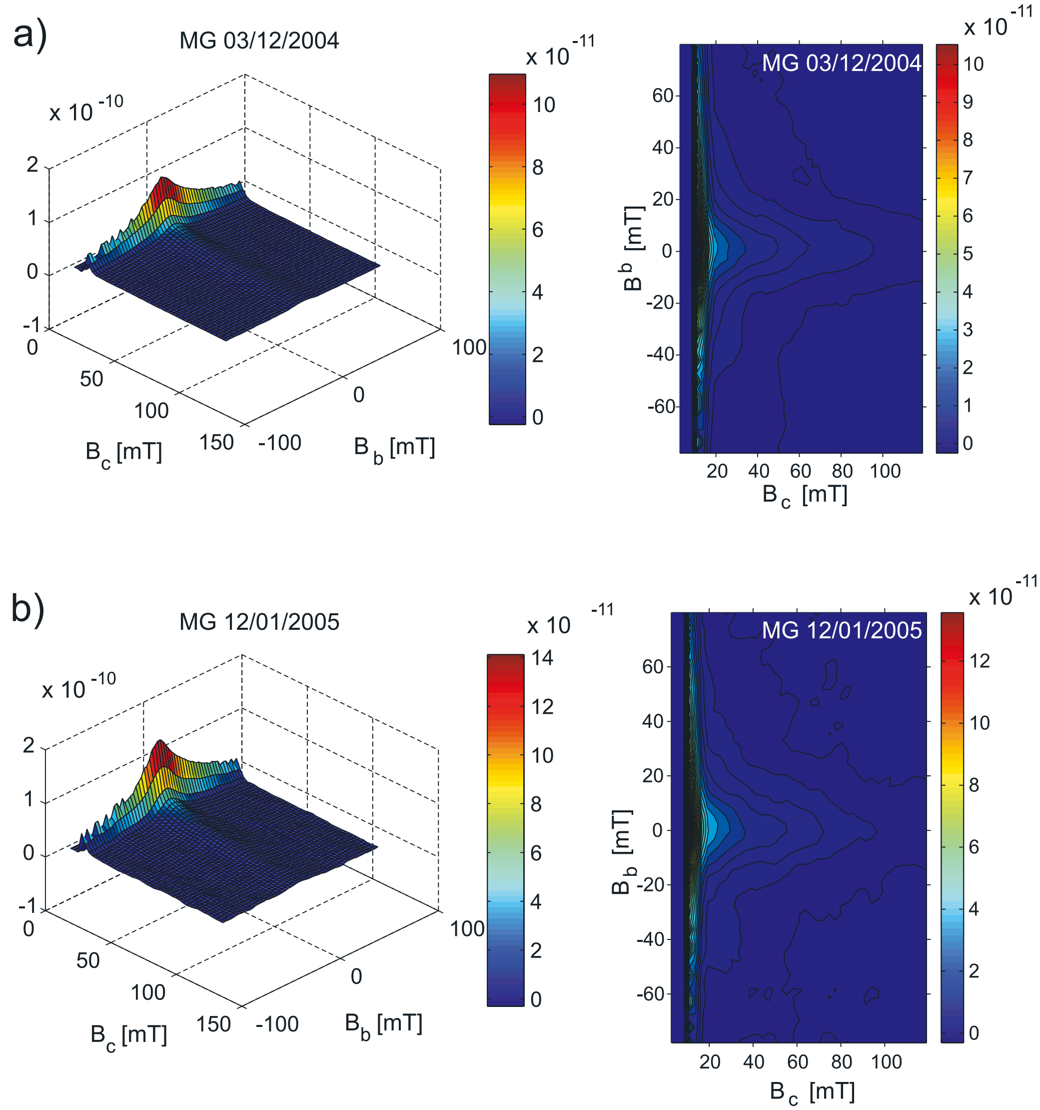

Figure 11. FORC distribution for samples (a) MG031204 representing the main input peak of dust from North Africa and (b) MG120105 representative of a high-pollution period. FORC diagrams have been computed, with a smoothing factor of 7, using the FORCobello software written by M. Winklhofer (available at http://venus.geophysik.uni-muenchen.de/~michael/forcnew/). By comparison with the interpretative framework of *Roberts et al.* [2000], these FORC distributions are consistent with a mixture of superparamagnetic and multidomain particles, with negligible interaction. The FORC distributions for the two samples are remarkably similar; we point out that the dramatic input of north African dust on 3 December 2004 (see also Figure 3) did not produce visible effects on the FORC data, indicating that magnetic properties of such dust are negligible.

[52] The component analysis of ARM demagnetization data clearly shows that the magnetic composition of all the analyzed sites, except Fontechiari, is fairly homogeneous and allows to conclude that the PM_{10} shares the same sources of magnetic minerals through the entire period of observation (Table 4).

[53] As a whole, the concentration of the various magnetic components is higher in polluted sites (Magnagrecia, Frosinone, see also Figure 8) and during periods of increased pollution (e.g., 5–14 January 2005), as expected from the magnetic susceptibility measurements. On the other hand, a clear systematic correlation between the magnetic composition of the samples and the degree of pollution could not be recognized. Indeed, the high-coercivity component (ARM3), accounted for 20–25% of the total ARM at all sites during 5–14 January 2005 (Table 5). This suggests a regional spreading of pollutants during periods of stagnant atmosphere.

[54] The magnetic minerals contained in dust collected in the Gubrist motorway tunnel are mainly SP: More than 80% of an IRM acquired at 4K was lost upon warming to room temperature. Measurements of the frequency dependence of susceptibility as a function of temperature suggests a broad distribution of blocking temperatures peaking at ≈140 K (Figure 12). A similar distribution of blocking temperatures, peaking at 100 K, was observed by *Gómez-Paccard et al.*

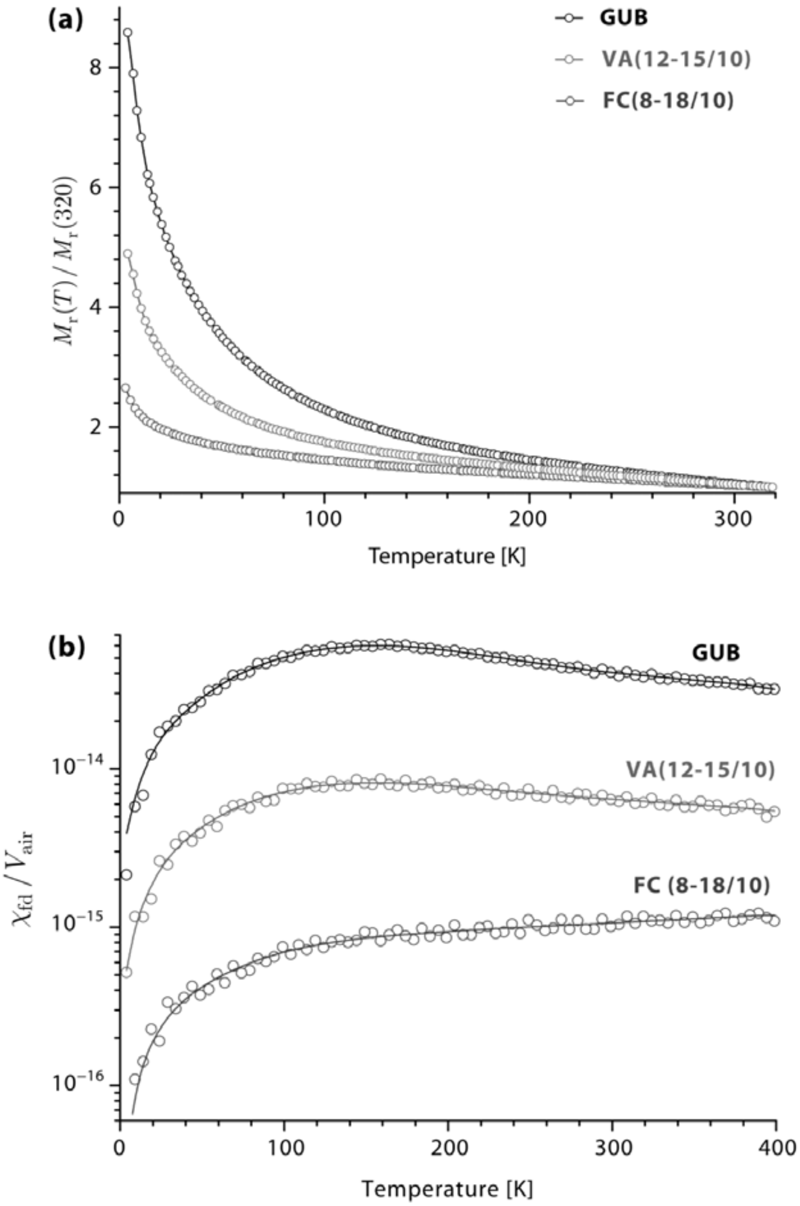

Figure 12. Low-temperature measurements for two representative specimens from Villa Ada (12–15 October 2005) and Fontechiari (8–18 October 2005) stations, together with sample GUB collected in the center of a motorway tunnel [*Spassov et al.*, 2004]. (a) Thermal demagnetization of an IRM imparted at 4 K in a 2.5 T field. The curves are normalized to the remanence measured at 320 K. (b) Frequency dependence of magnetic susceptibility measured at 4–400 K. The unitless χ_{fd} is obtained by normalizing the measured susceptibility (magnetic moment/applied field, m^3) by the volume of filtered air (23.472 m^3/d multiplied by the number of sampled days).

[2004] for PM collected in Madrid and linked to the presence of highly oxidized magnetite or maghemite. The pollution-related component ARM3 identified in several samples of the present study is highly viscous: Experiment shows that the relative contribution of this component to the total ARM depends on the time elapsed between ARM acquisition and measurements. This component is also characterized by a unusually high MDF of about 100 mT, which is well constrained (DP $\approx$ 2), suggesting a fairly homogeneous source. *Flanders* [1999] reports that coercivity values of fly ash collected near fossil fuel power stations are a factor 2–3 higher than those typical of soil erosion minerals. He interpreted the enhanced coercivity as arising from the high mechanical stress developed in rapidly cooled ash particles.

[55] The Day and FORC diagrams (Figures 10 and 11) indicate also the additional presence of grains with a MD behavior in the pollution component. We tentatively interpret this MD fraction as the carrier of the low-coercivity component ARM1 (MDF = 10–12 mT). The origin of this magnetic fraction is not clear; it could be related to the

presence of metallic fragments released by brakes and other abraded parts of vehicles.

6. Conclusions

[56] This study pointed out that an empirical linear correlation can be found between the magnetic properties of daily filters in air-monitoring stations (i.e., their low-field magnetic susceptibility, *k*) and the PM_{10} concentration. The correlation is valid at a local scale in absence of significant exogenous PM_{10} inputs and may be used to estimate the amount of the latter during "perturbed" days. The correlation coefficients are different at each station and variation of local conditions may result in a permanent change of the local empirical linear correlation linking *k* to PM_{10} concentration (as shown by the Latina station, see Figure 5).

[57] The abundance of magnetic particles in the total PM_{10} is estimated by the mass-specific low-field magnetic susceptibility (χ), which may be computed from the *k* and PM_{10} concentration values. The χ values obtained in this study indicate that the concentration of magnetic particles in the total PM_{10} collected in the larger urban setting (i.e., Rome, Magnagrecia and Villa Ada stations) is 2 orders of magnitude higher than in the rural background station (Fontechiari), and distinctly higher than in provincial towns (Viterbo, Latina). The industrial area of Frosinone scalo shows χ values comparable with those measured in the large green park of Villa Ada in Rome, but distinctly lower than those obtained in the high-traffic Via Magnagrecia.

[58] Magnetic particles in the PM_{10} of the Latium Region are mainly composed of low-coercivity, magnetite-like, minerals. This is consistent with former reports from other European metropolis (i.e., Münich [see *Muxworthy et al.*, 2002, 2003], Zürich [see *Spassov et al.*, 2004], Madrid [see *Gómez-Paccard et al.*, 2004]). The ARM demagnetization analysis indicate that the whole assemblage of magnetic particles found in PM_{10} at all Latium stations, except the rural background (Fontechiari), consists of three components with different coercivity distributions (ARM1 with MDF $\approx$ 10 mT; ARM2 with MDF $\approx$ 45 mT and ARM3 with MDF $\approx$ 100 mT). During periods of regional high pollution these components may be recognized at Fontechiari too.

[59] A detailed rock magnetic analysis of selected PM_{10} samples indicates a mixture of ferrimagnetic minerals that may be associated to distinct natural and anthropogenic sources. Hysteresis properties of PM_{10} collected at the rural background site (Fontechiari) during a period of low pollution are not distinguishable from those of typical eolian deposits. These properties indicate PSD ferrimagnetic carriers that we therefore identify as the magnetic content of natural dust. This magnetic fraction can be related to coercivity component ARM2.

[60] The mixing trend of hysteresis ratios obtained for selected samples from various environmental conditions suggests that the PM_{10} samples are a mixture of PSD grains (natural dust) and SP and MD grains (pollution). Though the exact mineralogy of the pollution component is unknown, rock magnetic data clearly show that the magnetic minerals contained in dust produced by vehicular traffic are mainly SP. We tentatively interpret the SP fraction as a product of rapidly cooling motor combustion, and the MD fraction (the likely carrier of the ARM1 component) as a product of the abrasion of metallic parts of the vehicles.

[61] **Acknowledgments.** This study was undertaken in the framework of a cooperative Project between INGV, Regione Lazio and ARPA Lazio and was funded by Regione Lazio. We are grateful to Angelo Lozito for having stimulated this research; this work benefited from many fruitful discussions with him and Giorgio Catenacci. We thank Francesco Troiano and Andrea Bolignano for continuous assistance and support with the data on PM_{10} concentration. Thoughtful reviews by Eduard Petrovsky and Adrian Muxworthy allowed us to greatly improve the manuscript.

References

Avila, A., I. Queralt-Mitjans, and M. Alarcon (1997), Mineralogical composition of African dust delivered by red rains over northeastern Spain, *J. Geophys. Res.*, *102*, 21,977–21,996.

Caquineau, S., A. Gaudichet, L. Gomes, and M. Legrand (2002), Mineralogy of Saharan dust transported over northwestern tropical Atlantic Ocean in relation to source regions, *J. Geophys. Res.*, *107*(D15), 4251, doi:10.1029/2000JD000247.

Cisowski, S. (1981), Interacting vs. non-interacting single-domain behavior in natural and synthetic samples, *Phys. Earth Planet. Inter.*, *26*, 77–83.

Day, R., M. D. Fuller, and V. A. Schmidt (1977), Hysteresis properties of titanomagnetites: Grain size and composition dependence, *Phys. Earth Planet. Inter.*, *13*, 260–266.

Dunlop, D. J. (2002a), Theory and application of the Day plot (M_{rs}/M_s versus H_{cr}/H_c): 1. Theoretical curves and tests using titanomagnetite data, *J. Geophys. Res.*, *107*(B3), 2056, doi:10.1029/2001JB000486.

Dunlop, D. J. (2002b), Theory and application of the Day plot (M_{rs}/M_s versus H_{cr}/H_c): 2. Application to data for rocks, sediments, and soils, *J. Geophys. Res.*, *107*(B3), 2057, doi:10.1029/2001JB000487.

Egli, R. (2003), Analysis of the field dependence of remanent magnetization curves, *J. Geophys. Res.*, *108*(B2), 2081, doi:10.1029/2002JB002023.

Egli, R. (2004), Characterization of individual rock magnetic components by analysis of remanence curves, 1. Unmixing natural sediments, *Stud. Geophys. Geod.*, *48*, 391–446.

Egli, R., and W. Lowrie (2002), Anhysteretic remanent magnetization of fine magnetic particles, *J. Geophys. Res.*, *107*(B10), 2209, doi:10.1029/2001JB000671.

Flanders, P. (1999), Identifying fly ash at a distance from fossil fuel power stations, *Environ. Sci. Technol.*, *33*, 528–532.

Flanders, P. J. (1994), Collection, measurement, and analysis of airborne magnetic particulates from pollution in the environment, *J. Appl. Phys.*, *75*, 5931–5936.

Galassi, C., B. Ostro, F. Forastiere, S. Cattani, M. Martuzzi, and R. Bertolini (2000), Exposure to PM_{10} in the eight major Italian cities and quantification of the health effects, poster presented at the ISEE 2000 Meeting, Buffalo, N. Y., 19–22 Aug. (Available online at http://www.who.dk/document/hms/pm10.pdf)

Gautam, P., U. Blaha, E. Appel, and G. Neupane (2004), Environmental magnetic approach towards the quantification of pollution in Kathmandu urban area, Nepal, *Phys. Chem. Earth*, *29*, 973–984.

Georgeaud, V. M., P. Rochette, J. P. Ambrosi, D. Vandamme, and D. Williamson (1997), Relationship between heavy metals and magnetic properties in a large polluted catchments: The Etang de Berre (south France), *Phys. Chem. Earth*, *22*, 211–214.

Goddu, S. R., E. Appel, D. Jordanova, and F. Wehland (2004), Magnetic properties of road dust from Visakhapatnam (India)–relationship to industrial pollution and road traffic, *Phys. Chem. Earth*, *29*, 985–995.

Gómez-Paccard, M., G. McIntosh, V. Villasante, M. L. Osete, J. Rodriguez-Fernández, and J. C. Gómez-Sal (2004), Low-temperature and high magnetic field measurements of atmospheric particulate matter, *J. Magn. Magn. Mater.*, *272*, 2420–2421.

Hanesch, M., R. Scholger, and D. Rey (2003), Mapping dust distribution around an industrial site by measuring magnetic parameters of tree leaves, *Atmos. Environ.*, *37*, 5125–5133.

Harrison, R. M., and J. Yin (2000), Particulate matter in the atmosphere: Which particle properties are important for its effect on health?, *Sci. Total Environ.*, *249*, 85–101.

Hoffmann, V., M. Knab, and E. Appel (1999), Magnetic susceptibility mapping of roadside pollution, *J. Geochem. Explor.*, *66*, 313–326.

Hunt, A., J. Jones, and F. Oldfield (1984), Magnetic measurements and heavy metals in atmospheric particulates of anthropogenic origin, *Sci. Total Environ.*, *33*, 129–139.

Liu, Q., M. J. Jackson, S. K. Banerjee, B. A. Maher, C. Deng, Y. Pan, and R. Zhu (2004a), Mechanism of the magnetic susceptibility enhancements of the Chinese loess, *J. Geophys. Res.*, *109*, B12107, doi:10.1029/2004JB003249.

Liu, Q., S. K. Banerjee, M. J. Jackson, C. Deng, Y. Pan, and Z. Rixiang (2004b), New insights into partial oxidation model of magnetites and thermal alteration of magnetic mineralogy of the Chinese loess in air, *Geophys. J. Int.*, *158*, 506–514.

Maher, B. A., R. Thompson, and M. W. Hounslow (1999), Introduction, in *Quaternary Climates, Environments and Magnetism*, edited by B. A. Maher and R. Thompson, pp. 1–48, Cambridge Univ. Press, New York.

Matzka, J., and B. A. Maher (1999), Magnetic biomonitoring of roadside tree leaves: Identification of spatial and temporal variations in vehicle-derived particles, *Atmos. Environ.*, *33*, 4565–4569.

Moreno, E., L. Sagnotti, A. Winkler, J. Dinarès-Turell, and A. Cascella (2003), Biomonitoring of traffic air pollution in Rome using magnetic properties of tree leaves, *Atmos. Environ.*, *37*, 2967–2977.

Morris, W. A., J. K. Versteeg, D. W. Bryant, A. E. Legzdins, B. E. McCarry, and H. X. Marvin (1995), Preliminary comparisons between mutagenic and magnetic susceptibility of respirable airborne particle, *Atmos. Environ.*, *29*, 3441–3450.

Muxworthy, A. R., J. Matzka, and N. Petersen (2001), Comparison of magnetic parameters of urban atmospheric particulate matter with pollution and meteorological data, *Atmos. Environ.*, *35*, 4379–4386.

Muxworthy, A. R., E. Schmidbauer, and N. Petersen (2002), Magnetic properties and Mössbauer spectra of urban atmospheric particulate matter: A case study from Munich, Germany, *Geophys. J. Int.*, *150*, 558–570.

Muxworthy, A. R., J. Matzka, A. F. Davila, and N. Petersen (2003), Magnetic signature of daily sampled urban atmospheric particles, *Atmos. Environ.*, *37*, 4163–4169.

Pike, C. R., A. P. Roberts, and K. L. Verosub (1999), Characterizing interactions in fine magnetic particle systems using first order reversal curves, *J. Appl. Phys.*, *85*, 6660–6667.

Pope, C. A., III, and D. W. Dockery (1999), Epidemiology of particle effects, in *Air Pollution and Health*, edited by S. T. Holgate et al., pp. 673–705, Elsevier, New York.

Regione Lazio (2005), Progetto Polveri Sottili, Relazione di sintesi, stato di avanzamento al febbraio 2005, 56 pp., Rome, Feb.

Regione Lazio (2006), Progetto Polveri Sottili, final report, 266 pp., Rome, Jan.

Roberts, A. P., C. R. Pike, and K. L. Verosub (2000), FORC diagrams: A new tool for characterizing the magnetic properties of natural samples, *J. Geophys. Res.*, *105*, 28,461–28,475.

Sagnotti, L., P. Rochette, M. Jackson, F. Vadeboin, J. Dinarès-Turell, A. Winkler, and ''Magnet'' Science Team (2003), Inter-laboratory calibration of low field and anhysteretic susceptibility measurements, *Phys. Earth Planet. Inter.*, *138*, 25–38.

Shilton, V. F., C. A. Booth, J. P. Smith, P. Giess, D. J. Mitchell, and C. D. Williams (2005), Magnetic properties of urban street dust and their relationship with organic matter content in the West Midlands, UK, *Atmos. Environ.*, *39*, 3651–3659.

Shu, J., J. A. Dearing, A. P. Morse, L. Z. Yu, and N. Yuan (2001), Determining the sources of atmospheric particles in Shanghai, China, from magnetic and geochemical properties, *Atmos. Environ.*, *35*, 2615–2625.

Spassov, S., F. Heller, R. Kretzschmar, M. E. Evans, L. P. Yue, and D. K. Nourgaliev (2003), Detrital and pedogenic mineral phases in loess/paleosol sequence at Lingtai (Central Chinese Loess Plateau), *Phys. Earth Planet. Inter.*, *140*, 255–275.

Spassov, S., R. Egli, F. Heller, D. K. Nourgaliev, and J. Hannam (2004), Magnetic quantification of urban pollution sources in atmospheric particulate matter, *Geophys. J. Int.*, *159*, 555–564.

Urbat, M., E. Lehndorff, and L. Schwark (2004), Biomonitoring of air quality in Cologne conurbation using pine needles as a passive sampler–part I: Magnetic properties, *Atmos. Environ.*, *38*, 3781–3792.

Weber, S., P. Hoffmann, J. Ensling, A. N. Dedik, S. Weinbruch, G. Miehe, P. Gutlich, and H. M. Ortner (2000), Characterization of iron compounds from urban and rural aerosol sources, *J. Aerosol Sci.*, *31*(8), 987–997.

Wichmann, H. E., and A. Peters (2000), Epidemiological evidence of the effects of ultrafine particle exposure, *Philos. Trans. R. Soc. London, Ser. A*, *358*, 2751–2769.

World Health Organization (2002), Health impact assessment of air pollution in the eight major Italian cities, EURO/02/5040650, Geneva. (Available at http://www.euro.who.int/document/e75492.pdf)

World Health Organization (2004), Particulate matter air pollution: How it harms health, Fact Sheet EURO/04/05, Berlin, Copenhagen, Rome, 14 April. (Available at http://www.euro.who.int/document/mediacentre/fs0405e.pdf)

Xie, S., J. A. Dearing, and J. Bloemendal (2000), The organic content of street dust in Liverpool, UK, and its association with dust magnetic properties, *Atmos. Environ.*, *34*, 269–275.

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