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In-situ physical and chemical characterization of the Eyjafjallajökull aerosol plume in the free troposphere over Italy

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Abstract

Continuous measurements of physical and chemical properties at the Mt. Cimone GAW-WMO Global Station (2165 m a.s.l.) allowed the detection of the volcanic aerosol plume resulting from the Eyjafjallajökull eruption of spring 2010. The event affected the site after a transport over a distance of more than 3000 km. Two main transport episodes were detected during the eruption period, showing a volcanic fingerprint discernible against the free tropospheric background conditions typical of the site, the first from 19 to 21 April and the second from 18 to 20 May 2010. The paper reports the modification of aerosol characteristics observed during the two episodes, both characterized by an abrupt increase in fine and, especially, coarse mode particle number.

Analysis of major, minor and trace elements by different analytical techniques (Ionic Chromatography, PIXE-PIGE and ICP-MS) were performed on aerosols collected by ground level discrete sampling. The resulting database allows the characterization of aerosol chemical composition during the volcanic plume transport and in background conditions. During the passage of the volcanic plume, the fine fraction was dominated by sulphates, denoting the secondary origin of this mode, mainly resulting from in-plume oxidation of volcanic SO_2. By contrast, the coarse fraction was characterized by increased concentration of numerous elements of crustal origin, such as Fe, Ti, Mn, Ca, Na, and Mg, which enter the composition of silicate minerals. Data analysis of selected elements (Ti, Al, Fe, Mn) allowed the estimation of the volcanic plume's contribution to total PM_{10}, resulting in a local enhancement of up to 9.5 $\mu g m^{-3}$, i.e. 40 % of total PM_{10}, on 18 May, which was the most intense of the two episodes. These results appear significant, especially in the light of the huge distance of Mt. Cimone from the source, confirming the widespread diffusion of the Eyjafjallajokull ashes over Europe.

1 Introduction

Volcanoes are highly active sources of aerosols and gases, hence having an impact on the atmospheric radiative budget. The effects can be both direct, by backscattering and absorption of solar radiation, as well as indirect, by increasing the condensation nuclei number and ultimately affecting cloud properties and lifetime. Volcanic emissions mainly consist of two components: primary solid and liquid particles, forming through magma disintegration and erosion of the conduit wall, and gaseous compounds (SO_2, H_2S, HCl and HF), which are potential precursors of secondary aerosol, mainly represented by sulphate particles produced by gas- or cloud-phase oxidation of SO_2. Volcanic aerosol generally exhibits multi-mode size distributions, due to its multiple formation processes, with fragmented magma and erosion of particles from the vent walls in the coarse fraction (> 1 µm), and finer particles resulting from the condensation of volatiles and gas-phase reaction products in the accumulation mode (Mather et al., 2003; Mather, 2008).

Volcanic particles can undergo long-range transport in the atmosphere, during which their size distribution evolves due to the settling of the largest particles and condensational growth of particles in the fine and ultrafine fractions. In addition, the aerosol residence in the atmosphere of several days modifies the original chemical composition following in-plume chemical processing. Hence, the longer the distance from the source, the more difficult it is to distinguish an unequivocal volcanic signature. Eventually, the particle concentration decreases until it is no longer distinguishable from the background atmospheric concentration. At long distances from the source, volcanic plumes can be better identified at high-altitude sites, where the masking effect of background particles forming in the planetary boundary layer (PBL) is reduced.

Research on volcanic aerosol was in the past mainly oriented towards coarse particles, while the fine fraction received less attention. Currently, however, interest is focusing on the joint investigation of gases, fine aerosols and coarse aerosols, in order to understand the spatial distribution of the different types of compounds produced by the eruption, and their transformation during transport, since a significant part takes the form of fine particles, which are responsible for severe health effects (Pope, 2000).

With its tropospheric lifetime of several days, SO_2 is an excellent tracer of long-range transport of volcanic emissions, especially in areas not strongly influenced by anthropogenic emissions (Flentje et al., 2010). During transport, sulphuric acid is formed by gas-phase and aqueous-phase reactions and by heterogeneous reactions on the surface of solids (Mather et al., 2006). H_2SO_4 is taken up very efficiently by pre-existing aerosol particles and cloud droplets. Alternatively, it can nucleate to form new particles, especially in conditions of low condensation sinks (von Glasow, 2009). Volcanic plumes are therefore normally characterized by increased concentrations of gaseous SO_2 and sulphate, a chemical feature that is sometimes observed even at long distances, despite the widespread presence of anthropogenic emissions. Super-micrometric particle number concentrations in the presence of volcanic plumes are generally enhanced due to primary ash particles (coarse fraction). Occasionally, new-particle formation in the ultrafine size range also occurs in volcanic plumes (Flentje et al., 2010).

Volcanic aerosols are considerably enriched in many trace elements, including alkali, alkali-earth, transition, and heavy metals, which can either originate via magma volatilization or (in the case of soluble species) from acid leaching of the vent wall rock or silicate ash particles (Mather et al., 2003). Aerosols within volcanic plumes may be highly enriched in these elements relatively to either crustal material or ash deposited in the immediate vicinity of the eruption (Symonds et al., 1987).

Apart from cataclysmal eruptions, which affect climate on a global scale, moderate eruptions and/or quiescent degassing can have considerable local effects. However, their impact is spatially and temporally inhomogeneous, given the turbulent nature of the lower troposphere, where residence times would typically be less than 1 week.

The Icelandic volcano Eyjafjallajökull (63°38′ N, 19°36′, 1660 m a.s.l.) erupted on 20 March 2010, after a relatively silent period of nearly 190 yr. A major outbreak of the central crater under the covering ice cap started on 14 April and continued until 24 May, ejecting a very large amount of gases and fine ashes into the atmosphere.

The eruption was considered of moderate magnitude, but emissions remained in the troposphere (up to 10 km) for days, and affected highly populated Central Europe, inducing disruption of air traffic for several days. Meanwhile, several ground-based instruments and stations were able to monitor the volcanic plume, representing a unique opportunity for the study of aged volcanic aerosol.

The dynamics of the eruption have been extensively described in the literature in terms of ground-based remote sensing and satellite observations, highlighting the optical properties of the volcanic plume (e.g., Pappalardo et al., 2012; Ansmann et al., 2010; Flentje et al., 2010; Madonna et al., 2010; Mona et al., 2010; Schumann et al., 2011; Stohl et al., 2011; Schäfer et al., 2011). However, the chemical composition of aerosol from airborne measurements (Schumann et al., 2011; Bukowiecki et al., 2011; Rauthe-Schöch et al., 2012; Andersson et al., 2013) or from ground level samplings (Revuelta et al., 2012; Schleicher et al., 2012; Lettino et al., 2012; Beeston et al., 2012; Bukowiecki et al., 2011; Colette et al., 2011) were derived in a few cases only.

Owing to its remoteness from intense pollution sources, Mt. Cimone (see site description below) offers a unique opportunity to observe the influence of airborne particulate sources, usually overshadowed by local sources at ground-level locations. A comprehensive suite of aerosol measurements was performed at this site during the eruption period. Such measurements, which allowed the detection of the volcanic plume transit a first time from 19 April to 21, and a second time between 18 and 20 May 2010, therefore add some unique observations to the multitude of different measurements performed in various parts of Europe during the eruption.

The objective of this study is twofold: (1) to characterize the long-range transported volcanic plume that reached the station of Mt. Cimone from 19 April to 20 May 2010, through the continuous monitoring of physical parameters (size distribution, scattering, absorption, particle mass) and off-line chemical analysis; (2) to quantify the actual contribution of volcanic particles on the whole local aerosol burden, at a distance of more than 3000 km from the eruption, and to provide a review of similar measurements obtained in other parts of Europe from ground-based stations and from model calculations.

2 Experimental

2.1 Sampling site

Mt. Cimone (CMN) is the highest summit of the Italian Northern Apennines (44.11° N, 10.42° E, 2165 m a.s.l.). This sampling site, with its 360° free horizon, can be considered for most of the year representative of the southern Europe free troposphere (Bonasoni et al., 2000; Fischer et al., 2003). Only during fair weather days in the warm months, CMN is influenced by thermally induced injections of pollutants from the PBL (Cristofanelli et al., 2009). CMN therefore constitutes a suitable location for investigating the influence of long-range transport of air masses which are potentially able to modify the tropospheric background conditions of the Mediterranean basin (Marinoni et al., 2008).

2.2 Monitoring of aerosol physical parameters

In this work we analyse the aerosol particle size distribution, equivalent BC concentration and scattering coefficient at 525 nm, together with meteorological parameters (wind speed and direction) and surface ozone concentrations, continuously monitored at CMN.

The Multi Angle Absorption Photometer (MAAP, Thermo Scientific) provides equivalent black carbon concentrations (eq BC) by measuring the aerosol absorption coefficient σ_{abs} at 637 nm (Petzold et al., 2004). The scattering coefficient σ_{sca} at 525 nm is measured by a M9003 integrating nephelometer (ECOTECH) whose data are adjusted to standard temperature and pressure conditions (0 °C; 1000 hPa). Fine particle concentrations are measured by a Thermo Systems Inc. (TSI) Condensation Particle Counter (CPC) 3772, which detects airborne particles with diameter ranging from

10 nm (counting efficiency > 50 %) to 3 µm, over a concentration range from 0 to 104 particles cm^{-3}.

For the continuous size distribution measurements, an optical particle counter (OPC) and a differential mobility particle sizer (DMPS) are deployed, both connected to a total aerosol sampling head. The OPC (Mod. GRIMM 1.108) measures concentration and size distribution of particles with optical diameter between 0.30 and 20 µm in 15-size bins. According to the specifications, the reproducibility of the OPC in particle counting is ±2 % (Putaud et al., 2004). These measurements permit the determination of a fine (0.3 µm $\leq D_p \leq$ 1 µm) and coarse mode (1 µm $\leq D_p \leq$ 20 µm) aerosol fraction. As described in detail by Bukowiecki et al. (2011), aerosol measurements performed with a Polystyrene Latex (PSL)-calibrated OPC will result in a diameter shift of the actual size distribution due to the different refractive indices of volcanic ash. This results in a larger uncertainty in OPC measurements. The DMPS measurements were within 10 % for the size range 20–200 nm against a reference system under controlled laboratory conditions, while for sizes up to 500 nm the deviation was up to 25 % (Wiedensohler et al., 2010).

2.3 Chemical analyses

Within the EUSAAR and the ACTRIS EC Projects, background aerosol chemical composition has been continuously measured at CMN since February 2009. Samples for off-line chemical analyses are collected on quartz fibre filters by a dichotomous virtual impactor – Model 310 Universal Air Sampler TM, which allows the collection of the fine (PM_1) and coarse (PM_1–PM_{10}) fraction of particulate matter at a flow rate of 300 L min^{-1}. Except for special intensive field campaigns, such as the one described in this paper, one nocturnal sample is collected per week (from 10.00 p.m. to 06.00 a.m. LT, 8 h sampling time). The sample is analysed for total carbon (TC), and water-soluble organic carbon (WSOC) by an Analytik-Jena multianalyzer N/C (model 2100), equipped with a non-dispersive infrared (NDIR) detector, while ionic chromatography is employed for major water-soluble inorganic anions (Cl^-; NO_3^-; SO_4^{2-}), cations

(Na^+; NH_4^+; K^+; Mg^{2+}; Ca^{2+}) and organic acids. For this purpose two DIONEX ICS2000 are dedicated respectively to anions (Dionex IonPac AS11 separation column with IonPac AG11 guard column, mobile phase KOH autogenerated with gradient program) and cations (Dionex IonPac AS16 separation column with IonPac AG16 guard column, mobile phase Metansolphonic Acid autogenerated with isocratic elution).

The choice of nocturnal sampling is oriented towards catching the free troposphere conditions both in the cold and in the warm season, because occasionally, especially in the central hours of summer days, the site experiences maxima in aerosol number and black carbon (BC) concentrations due to enhanced vertical transport from lower elevations and mountain breeze wind regime (Cristofanelli et al., 2007; Marinoni et al., 2008; Carbone et al., 2010). Based on transport forecasts, indicating the possible arrival of the volcanic plume, from 20 April the routine sampling was intensified, with the collection of 11 samples (9 nocturnal and 2 diurnal) covering the period from 20 April to 19 May 2010.

As volcanic particulate emissions are primarily composed of non-carbonaceous and insoluble material, often characterized by the presence of trace elements normally not found in ambient aerosol, such filters were analysed for additional chemical parameters: the total content of elements Na, Mg, Al, S, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Br, Sr, Y, Zr, Mo and Pb was determined through quantitative multi-elemental analysis by Particle Induced X-ray Emission-Particle Induced Gamma-ray Emission (PIXE-PIGE). PIXE-PIGE analysis was performed at the 3 MV Tandetron accelerator in Florence, with a 5 nA 3 MeV proton beam using the external beam set-up described in Calzolai et al. (2006). This technique represents a very powerful tool for environmental and geological studies and has been successfully applied in the characterization of volcanic emissions (Koltay et al., 1999). It provides multi-elemental analysis (elements with atomic number larger than 10) of samples with high sensitivity and non-destructivity, hence allowing the subsequent application of other techniques.

A series of metals and metalloids (Li, B, Na, Mg, Al, P, Ca, Cr, Mn, Co, As, Mo, Cd, Sb, Cs, Ba, La, Ce, W, Tl, Pb and U) extracted by acetate buffer solution at pH

4.5 was analysed by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) (820 Varian, Australia), according to the method proposed by Canepari et al. (2006). This soluble fraction, compared to the total element determination, provides more information about the environmental and biological availability of the element and allows obtaining a higher selectivity as source tracer (Canepari et al., 2010), the soluble fraction being mostly associated to combustion sources, and the insoluble one to crustal contributions. Finally, organic carbon (OC) and elemental carbon (EC) were analysed by a thermo-optical transmission analyzer (Sunset Laboratory Inc., OR-USA) using a NIOSH-Quartz thermal protocol (Perrino et al., 2007).

Since filters are subjected to both water soluble organic carbon and inorganic ions determination throughout the year, measurements of these parameters before and after the volcanic transport events were available for comparison with the collected volcanic aerosol plume. Trace elements from PIXE-PIGE and ICP-OES, instead represent quite new data for CMN. Hence no comparisons with background conditions at the same site are possible, with the exception of some measurements performed by PIXE-PIGE in August 2007 during a Saharan dust transport episode (Cristofanelli et al., 2009). Other measurements of metals and trace elements had been performed during a summer campaign in 2004 by XRF (Marenco et al., 2006), and these data can be used as reference, although season and analytical techniques were different.

2.4 HYSPLIT 3-D back-trajectories

144 h 3-D air-mass back-trajectories ensembles were calculated using the HYSPLIT model to describe the air-mass circulation and origin that affected CMN during the Eyjafjallajökull events (Draxler and Rolph, 2011). The model calculations were based on the Global Data Assimilation System (GDAS) meteorological field produced by the National center for Environmental Prediction (NCEP) with a horizontal resolution of $1^\circ \times 1^\circ$. For every point along the trajectory (time resolution: 1 h), the model provides the geographic location and altitude (or pressure level) of the air parcel. Sub-grid scale processes like convection and turbulent diffusion are not represented by HYSPLIT

back-trajectories. To compensate partially for these uncertainties, every 6 h starting from 1 April (00:00 UTC) to 30 May 2010 (00:00 UTC), an ensemble of seven back-trajectories was calculated, ending at the CMN location (±0.5° in latitude/longitude and ±250 m in altitude).

3 Results and discussion

3.1 Plume transport towards northern Italy and data overview

The Eyjafjallajökull eruption entered its explosive phase on 14 April and continued until 24 May 2010, with two main phases of intense ash and gas emission, spanning from 14–21 April to 1–18 May (Stöhl et al., 2011). During the eruption, persistent north-westerly winds over Iceland favoured the transfer of the volcanic plume south-eastwards, resulting in the dispersion of ashes over a large part of Europe at about 4–9 km above ground (Emeis et al., 2011; O'Dowd et al., 2012).

Remote sensing systems already observed the plume in the free troposphere over Europe on 16 April at an altitude of 3–6 km, but only on the afternoon of 17 April did intense vertical mixing and advection favour the entrainment of the volcanic material down to the ground in southern Germany (Flentje et al., 2010). Subsequently, on reaching the southernmost part of Europe, the air mass was deflected both east and west, with its presence observed over the Alps (Bukowiecki et al., 2011; Schäfer et al., 2011; Trickl et al., 2012). The transport to the south during this first approach of the plume was almost completely blocked by the Alps and by a low-pressure zone over northern Italy (Pappalardo et al., 2012; Trickl et al., 2012). Thus, the air mass resided over Central Europe for several days, until 19 April, when a redistribution of aged volcanic aerosol occurred from west to east and from north to south, and the plume was observed over Italy for the first time (Madonna et al., 2010; Lettino et al., 2012; Rossini et al., 2012; Belosi et al., 2011; Campanelli et al., 2012; Perrone et al., 2012). These changes in weather patterns were also discernible at CMN, where wind direction turned from northerly (on 17 April) to southerly (on 18 April), and eventually to northerly (on 19 April) (Fig. 3).

New significant eruptions occurred on 4–9 May and 14–19 May. The first of these events mainly influenced western Europe, from Great Britain to the Iberian Peninsula (Pappalardo et al., 2012), while the second travelled over Ireland (O'Dowd et al., 2012) and Great Britain and reached Central Europe (Bukowiecki et al., 2011; Hervo et al., 2012). Volcanic layers were observed in the Central and Eastern Mediterranean area, including Greece and Turkey, in the days from 18 to 22 May (Balis et al., 2010; Papayannis et al., 2012).

The HYSPLIT back-trajectories were analysed on a daily basis to establish a possible source–receptor relationship between the Eyjafjallajökull eruption and the atmospheric compounds variability observed at CMN. In particular, for each day from 1 April to 31 May 2010, evaluations were made of the total number of back-trajectory points falling within the geographical box 62.5–65.5° N; −21.5–17.5° E, roughly corresponding to Iceland's location (hereinafter denoted as the "Iceland" box). The choice of this rather large geographical box was made in order to take into account the possible uncertainty associated with back-trajectory calculation (according to Stöhl et al. (1998), up to 20 % of the travel distance from the end-point). As suggested by this analysis (Fig. 1a), air-masses possibly originating from the Iceland region and thus affected by the Eyjafjallajökull eruption, were present at CMN on several days. In particular, two major episodes were probably related to the volcanic plume transport: 19–21 April and 18–20 May 2010. During these two events, data clearly show the modification of the typical background aerosol at the site, normally characterized by only few coarse mode particles per cm^3. In both cases, the instruments recorded an enhanced particle number concentration, with the coarse mode greatly above the normal background (Fig. 1), as well as an increase in the scattering and absorption coefficients (Figs. 3 and 4).

A further significant increase in particle number in the coarse and accumulation modes occurred on 5 May 2010, in the presence of air-masses from Iceland. However, on this day, air-masses originating from north Africa (roughly 10–35° N; 15° W–30° E) were also present at CMN, resulting in a superimposed contribution of Saharan mineral dust (Fig. 1a). Again, accumulation mode particles showed a marked increase on 26 April, but on this occasion no simultaneous build-up of coarse particles was observed. This was attributed to the transport of polluted air from the PBL, following a period of high pressure.

The air masses originating from Iceland that passed over CMN between April and May travelled for about 5 days over a distance of more than 3000 km. For the typical ash collected during the explosive phase of the eruption near the source, diameter peaked at approximately 500 µm, with at least 20 % of mass < 10 µm (Gislason et al., 2011), and was therefore susceptible to being transported over long distance. Far from the source, aircraft measurements over France between 19 and 22 April showed increased particle concentrations both in the submicronic (~ 100 nm) and in supermicronic (~ 2 µm) modes in the PBL (Hervo et al., 2012). The following section provides a detailed description of the modifications in the aerosol size distribution and chemical composition at CMN during the two abovementioned major transport events (Table 1).

3.2 Case 1: 19–21 April 2010

Starting from 05:00 (UTC + 1) on 19 April 2010 an increase in coarse particle concentration was detected, from 0.09 cm^{-3} to 1.28 cm^{-3} on 20 April, when the peak reached the maximum concentration. As shown in Fig. 3, the period of the aerosol peak concentration was mainly characterised by the presence of northerly winds (from NW to NE) at the measurement site. The effects of the volcanic plume transport on the coarse particle number at CMN persisted until 22 April. In parallel with the coarse aerosol particles, an increase in the submicron aerosol fraction was also observed: two maxima were recorded on 19 April at 14:00 (4543 cm^{-3}) and 20 April at 16:00 (3967 cm^{-3}). Such values are significantly higher than the typical concentrations usually observed at CMN (Marinoni et al., 2008; Asmi et al., 2011). Meanwhile, the aerosol absorption coefficient (30 min average) increased from a value below the instrumental detection limit to a maximum of 6 Mm^{-1}, while the scattering coefficient increased from less than

$20\,\mathrm{M\,m^{-1}}$ to a maximum of $80\,\mathrm{M\,m^{-1}}$ on 20–21 April (Fig. 3). It is worth noting that, albeit over different ranges, all these atmospheric compounds showed the same temporal behaviour, suggesting the hypothesis that common processes or sources determined the occurrence of the observed peaks. The volcanic influence on the air mass is supported by the analysis of the HYSPLIT back-trajectories, showing air masses coming from the Icelandic region with an average transport time of $104 \pm 26\,\mathrm{h}$ (Fig. 2). The volcanic signal during this transit was quite weak but still discernible in the atmospheric composition measurements at CMN, though the plume was no longer visible from METEOSAT SEVIRI satellite imagery after the dilution undergone during the transport (Schäfer et al., 2011).

The air masses travelled at a rather low pressure level ($< 700\,\mathrm{hPa}$) and experienced fast downward transport over continental Europe, where mixing with regional PBL air masses probably occurred before reaching CMN (Fig. 2). The increase in the number of particles against concentrations prior to plume arrival (taking 17 April as reference) occurred in different proportions for the three particle modes: the highest increase was observed in the coarse (> 200 times), and accumulation modes (12 times), while the fine particle number concentration underwent a threefold increase only. Hence, the two orders of magnitude increase of the aerosol coarse fraction was the main evidence of volcanic ash advection.

In order to obtain a comprehensive view of the aerosol size distribution OPC ($D_{\mathrm{p,\,opt}} > 300\,\mathrm{nm}$) and DMPS data ($10 < D_{\mathrm{p,\,mob}} < 500\,\mathrm{nm}$) were combined. Assuming particle sphericity, aerosol number distributions were converted to volume distribution ($\mathrm{d}V/\mathrm{dlog}(\mathrm{D}_p)$), applying a correction as in Khlystov et al. (2004). The size distributions observed during the two volcanic events are reported in Fig. 5, where they are compared to a clean-atmosphere situation just before the plume's arrival (17 April). On 20 April, a marked increase in accumulation mode particles was detected, peaking at $304\,\mathrm{nm}$. It was accompanied by the build-up of a coarse mode, which was almost invisible before air-mass arrival, peaking at about $2.5\,\mathrm{\mu m}$. The fine sulphate concentration was moderately high in this first observation ($2.1\,\mathrm{\mu g\,m^{-3}}$), about five times greater than the average value for the period April–May 2009–2011 ($0.41 \pm 0.90\,\mu g\,m^{-3}$) (Fig. 1b). It should be noted that the intensive sampling started on 20 April at 11.00 a.m., when the volcanic plume was already over CMN (left part of the plot in Fig. 1b); therefore, the above sulphate concentration could not represent the peak value.

During this episode, about 90 % of the PM_{10} sulphate was found in the fine (< 1 µm) fraction, with nitrates, ammonium and organic carbon (Fig. 1b) peaking in correspondence to the sulphate maximum, a feature typical of pollution aerosol. Similar behaviour was observed during the episode of 26 April: a strong increase in the accumulation mode particle number (Fig. 1a), not paired with a simultaneous increase in coarse particles, and characterized by high concentrations of NO_3^- and WSOC, revealed the anthropogenic origin of pollutants from the Po Valley. Here, in fact, a period of high pressure favoured the stagnation of air, with a consequent deterioration in air quality.

3.3 Case 2: 18–20 May 2010

According to the HYSPLIT analysis, air masses originating from the Icelandic region were present at CMN on 18–20 May 2010, when northerly winds continuously affected the measurement site (Fig. 4). In particular, starting from 18 May at 14:00, high values of particle concentrations (peak values of coarse and accumulation mode particles: 4.2 and 144.7 cm^{-3}, respectively) and scattering/absorption coefficients (peak values 105.6 Mm^{-1} and 5.5 Mm^{-1}, respectively) were observed at CMN. A sudden decrease in particle concentration and scattering/absorption coefficient occurred on 19 May at 09:45, when the measurement site was temporarily in cloud. It is conceivable that aerosol scavenging and cloud processing occurred in these conditions, which might explain the low concentrations of aerosol chemical constituents in the second sample collected during the May episode (Fig. 1b, grey bar).

A high coarse particle concentration ($> 1.0\,cm^{-3}$) was again observed from 19 May at 18:00 until 20 May at 13:00. During this period, an increase in fine particles (up to 6700 cm^{-3}) was also observed, indicating the presence of an air mass characterised by high aerosol loading. Absorption and scattering coefficients peaked at 4.1 $\mathrm{Mm^{-1}}$ and 134.5 $\mathrm{Mm^{-1}}$, respectively. The accumulation mode particles exhibited two abrupt concentration increases (up to 240 $\mathrm{cm^{-3}}$) on 20 May (at 3:15) and 21 May (at 4:15), in conditions of very high RH (100 %).

The coarse mode was more pronounced compared to the April event, with a peak concentration of 4.5 $\mathrm{cm^{-3}}$, corresponding to an increase of 900 times compared to the "background" concentration recorded on 17 April. The accumulation mode particles reached 150 $\mathrm{cm^{-3}}$, i.e. 30 times the concentration before the arrival of the volcanic plume (Fig. 4). The total number of fine particles was 6708 $\mathrm{cm^{-3}}$ (19 May at 20:00), 10 times higher than background conditions.

As suggested by the HYSPLIT analysis (Fig. 2), the atmospheric transport was more straightforward in this case than for the April episode, though it was characterised by a similar transport time (114 ± 14 h). The back-trajectory ensemble calculated for CMN spanned a large range of altitudes with air parcel pressure levels ranging from 900 to 400 hPa over the Iceland region, suggesting that the volcanic plume could have been intercepted by air masses even at a lower altitude, i.e. in proximity to the emission.

In contrast to the April event, research flights over the Irish Sea on 16 May, the Norwegian Sea on 19 May (Rauthe-Schöch et al., 2011), and over Switzerland and the Alps on 19 May (Bukowiecki et al., 2011) found the volcanic plume to be present in the free troposphere as a distinct layer which was only subsequently transported into the boundary layer south of the Alps. The aerosol volume distribution (Fig. 5) again showed the build-up of a coarse mode centred at 2.5 µm, 4 times higher than on 20 April, and an accumulation mode peaking at 234 nm, i.e. with a smaller size with respect to the previous episode (304 nm in May), but of comparable intensity.

In this occurrence, fine aerosol $\mathrm{SO_4^{2-}}$ reached a maximum of 3.5 $\mathrm{\mu g\,m^{-3}}$, more than eight times higher than the spring average for the years 2009–2011 (0.41 ± 0.90 $\mathrm{\mu g\,m^{-3}}$, 92th percentile of the spring data for the whole 3 yr dataset). Concentrations as high as 3.5 $\mathrm{\mu g\,m^{-3}}$, though infrequent, sometimes occur at CMN, especially during summer, when convection carries polluted air masses from the Po Valley to the station. What is unusual in this case, and provides the ash plume signature, is the absence of a simultaneous increase in organic or nitrate aerosol mass, as shown by the minimum of the NO_3^-/SO_4^{2-} ratio for the period (Fig. 1b). This ratio can be useful for distinguishing natural sources of sulphates from anthropogenic sources, where nitrates are generally simultaneously present. Volcanic plumes are normally characterized by high concentrations of sulphates not associated to either nitrate or organic compounds.

On 18 May an NH_4^+ concentration of 1.10 $\mu g\,m^{-3}$ was also measured. This was significantly higher than the spring average for the years 2009–2011 (0.15 ± 0.37 $\mu g\,m^{-3}$, 97th percentile of the whole 3 yr dataset) and indicates an enrichment in ammonium against background conditions, which are normally very low at CMN due to the absence of significant sources (Carbone et al., 2010). The simultaneous absence of an increased concentration of nitrates suggests that this ammonium could have resulted from the volcanic emission of NH_3 (Rose et al., 1986; Allen et al., 2000) or, more probably (given the long distance travelled by the air mass) from the neutralisation of acidic sulphate particles during the transport downwind.

In contrast to the April volcanic plume, on 18 May, 66 % of the total water soluble sulphate was found in the fine fraction, and the remaining 34 % in the coarse one (Fig. 1b), probably condensed on the surface of mineral particles, as confirmed by SEM observations performed by other authors (Lettino et al., 2012; O'Dowd et al., 2012). Lettino et al. (2012) undertook SEM analyses of particles collected during the volcanic plume in Rimini (150 km East of CMN).They observed in the coarse fraction the presence not only of mineral Ca-sulphates but also secondary particles morphologically similar to those constituting the fine fraction, i.e. complex secondary particles condensed on existing particles, mainly composed of ammonium nitrate and sulphate and sodium sulphate. Schumann et al. (2011) found the same fine secondary particles adhering to the surfaces of many coarse particles. In the presence of a large number of pre-existing ash particles, in fact, the energetically favoured path for gases is to condense on particle surfaces, rather than forming new ones through the nucleation of sulphuric acid droplets, as suggested by Schumann et al. (2011) and shown by Boulon et al. (2011). We argue that, in this process, the large number of coarse particles observed on 18 May, 3 times higher than in April (Fig. 4), competed with fine particles in the absorption of sulphuric acid and other volatile species. In both episodes chemical analysis showed a fine fraction of aerosol dominated by sulphate, and a coarse mode dominated by crustal elements, mainly Na, K, Al, Ca, Fe (here listed in order of decreasing concentration). This explains the bimodality of the volume distribution (Fig. 5), where the high concentration of coarse particles resulted from primary volcanic ash, while the accumulation mode was composed of liquid sulphuric acid or sulphate droplets, only partly neutralized by ammonia.

All physical and chemical data point to a substantial difference between the two volcanic plumes detected at CMN, which could be in part ascribed to changing eruption characteristics, but was mostly the result of different transport processes. The volcanic plume during the April event was more diluted and characterized by a pollution contamination, probably due to PBL interaction before reaching CMN: the PBL over the Po Basin was, in fact, well mixed on 20 April with a boundary layer height of about 1600 m (Belosi et al., 2011). In this case, aerosol at CMN also exhibited chemical features characteristic of anthropogenic PBL aerosol, i.e. high nitrates and organics, appearing as a mixture of volcanic and non-volcanic air (Fig. 1b). Moreover, elements typically attributed to anthropogenic emissions, such as Cd, Sb, Mo, Pb and Cr, were present in higher concentrations compared to the subsequent days. Such elements were mainly present in the fine and soluble fraction of PM, as typical of anthropogenic emissions (Canepari et al., 2010).

Further confirmation of the differences is the shift of the accumulation mode toward larger diameters in April (about 304 nm) compared to the May episode (234 nm) (Fig. 5), as similarly observed at Jungfraujoch (Switzerland) (Bukowiecki et al., 2011). Sulphate aerosol size, characterizing most of the accumulation mode, strongly depends on local meteorological conditions, particularly on relative humidity, which determines the hygroscopic growth of particles. For “non in-cloud” samplings in April and

May, RH values at CMN were quite similar (Figs. 3 and 4). Therefore the shift in diameter is likely to reflect the longer ageing of the April plume over Central Europe before being transported to CMN. Conversely, the coarse fraction diameter was centred at 2.5 µm in both cases, indicating distance from the source to be the main factor controlling the size distribution of this class of particles.

Because of the incomplete coverage by off-line chemical analyses of the April episode (sampling began when the plume was already over CMN), and because the May episode was much more impacted by ash particles than the previous one, the latter episode was felt to merit special attention. Thus, the following analysis based on trace element measurements refers to it alone.

3.4 Ash composition

Elemental analyses by PIXE-PIGE were performed only during the volcanic plume observation period, from 20 April to 19 May 2010, so no information exists on annual or monthly averages of such elements at CMN. The only data for elemental concentrations at this site are found in Marenco et al. (2006), where the aerosol chemical composition during summer 2004 was investigated by ED-XRF.

The present data show that, on 18 May, most of the typically crustal elements displayed high concentrations in the coarse fraction, reflecting their insoluble form as mineral crystals or glass shards. Major elements present in the coarse fraction of aerosol at CMN on 18 May were Na (1.05 $\mu g\,m^{-3}$), K (1.02 $\mu g\,m^{-3}$), Al (0.9 $\mu g\,m^{-3}$), Ca (0.65 $\mu g\,m^{-3}$), Fe (0.57 $\mu g\,m^{-3}$), Mg (0.26 $\mu g\,m^{-3}$), and Ti (0.073 $\mu g\,m^{-3}$), which match well with those defined by the Institute of Earth Sciences (http://earthice.hi.is/eyjafjallajokull_2010_chemical_composition) for the main oxides in the volcanic ash from Iceland.

In general, all the crustal elements had much higher concentrations during the volcanic plume transit than on other days in the same period, representing an important indication of the volcanic influence. However, this attribution is uncertain in the context of long-range transport of ubiquitous trace species (Colette et al., 2007). In fact, although some elements are enriched in the ash compared to the Earth's crust composition, they also have rather high background values due to anthropogenic emissions or other sources. In the study performed at CMN by Marenco et al. (2006), several elements among those enriched on 18 May (Mg, Al, Si, K, Ca, Ti, Mn, Fe and Sr) were associated to mineral dust transported from the northern Sahara and were found in the super-micronic fraction of aerosol. Saharan dust events are the main phenomena that usually contribute to the concentration of coarse particles at CMN, with maximum occurrence in spring-summer. Back trajectories (Figs. 1 and 2) had already ruled out a possible Saharan contribution to the high concentrations of crustal particles on 18 May, confirming the air mass passage over Iceland before reaching CMN. As further confirmation of the volcanic origin of these crustal particles, the total PM_{10} aerosol composition was compared to that of volcanic ash collected on 16 May (GSV165–3) at the eruption site (Fig. 6) (Sigmundsson et al., 2010; http://earthice.hi.is/eyjafjallajokull_2010_chemical_composition). The comparison was performed using elemental ratios, which are more robust than single element concentrations in univocally identifying sources.

A good agreement exists with the fallout sample for some elements (Ti, Mn, Mg, Sr). The larger deviations observed for other elements indicate that different sources could be present at CMN. In the case of more volatile elements (e.g. K), the enrichment may also be due to a fractionation effect subsequent to degassing and condensation on smaller particles (Witham et al., 2005), which undergo longer gravitational settling. The same enrichment probably occurred also for Zr, indicating that this element might have been abundant in the gas phase and condensed onto the ash particles, as also observed by Andersson et al. (2013).

ICP-MS analyses of the aerosol samples supplied further corroboration of the volcanic influence on the aerosol at CMN during the two episodes. They showed the enrichment of trace-elements such as the chalcophile metal Tl, whose presence in volcanic plumes is due to the degassing of silicate melts (Hinkley et al.,1994), and the more refractory rare earth elements La and Ce, known to be enriched in volcanic plumes (Moune et al., 2006).

3.5 PM_{10} mass budget

The chemical analyses of the PM_1 and PM_{1-10} samples collected on 18 May was used to reconstruct the collected PM_{10} mass. It was assumed that the crustal elements were present in their main oxide forms at the highest oxidation state.

Silicon, not determined due to the use of quartz sampling filters, was estimated from the detected Ti concentration, taking the Si/Ti ratio of the volcanic ash composition and transforming to SiO_2 mass. Sulphate, nitrate and ammonium concentrations were measured by ionic chromatography, while organic matter was estimated by multiplying the concentration of organic carbon by 1.8. This organic-mass-to-organic-carbon (OM/OC) ratio was derived from AMS data collected during the PEGASOS experimental campaign in summer 2012 at CMN. It is a value representative of oxygenated and/or functionalised organic species typical of processed aerosol (Gelencsér, 2004; Aiken et al., 2008), which takes into account the occurrence of biogenic aerosol and, in some cases, the presence of more oxygenated anthropogenic aged particles.

It is evident from the mass reconstruction (Fig. 7) that the fine fraction was dominated by ammonium sulphate (56 %). OC was also significant – 2.0 $\mu g\,m^{-3}$ organic matter (26 %) – while minor constituents were nitrate, EC and crustal oxides (0.84 $\mu g\,m^{-3}$). On average, during the whole observation period (20 April–19 May), the two main contributors, ammonium sulphate and organic matter, were found in a nearly 1 : 1 proportion, as also observed by Marenco et al. (2006) at the same site. However, the strong increase in fine sulphate in the presence of the volcanic plume made this component dominant on 18 May. By contrast, on the same day, PM_{1-10} was dominated by a mineral component represented by volcanic ash, amounting to 84 % of the total mass, with sulphates and organic matter as minor components.

The total reconstructed PM_{10} mass of aerosol during the 18 May volcanic transport amounted to 23 $\mu g\,m^{-3}$, of which 7 $\mu g\,m^{-3}$ as PM_1 and 16 $\mu g\,m^{-3}$ as PM_{1-10}, with a PM_1/PM_{10} ratio of 0.3. The same ratio measured gravimetrically at CMN during summer with 24 h samplings (Marenco et al., 2006) was 0.47. The significantly lower value during the volcanic transport is mostly attributable to the large increase in the coarse particles number occurring on 18 May resulting from the presence of the volcanic ash, which accounted for most of the aerosol mass.

3.6 Mass contribution of volcanic ash to total PM_{10} at CMN

In order to estimate the volcanic ash contribution on the total PM_{10} at CMN, the elemental composition of the filter samples was used, in particular the concentrations of some elements, namely Al, Fe, Ti and Mn. The same method was applied at sampling sites in other parts of Europe (Colette et al., 2011; Bukowiecki et al., 2011), in some cases using different tracers, such as Sr, Y and Zr (Krabbe et al., 2010; Schäfer et al., 2011).

Al, Fe, Ti and Mn were selected because they displayed a strong mutual correlation (correlation coefficients all > 0.9), confirming their common origin. They showed a simultaneous increase and subsequent decrease during the episodes of 20 April and 18 May, which probably indicates they share the same origin. Moreover, these species are known to be important contributors to volcanic ash chemical composition, with abundances of 8.3 % (Al), 7.5 % (Fe), 1.0 % (Ti) and 0.2 % (Mn) (http://earthice.hi.is/eyjafjallajokull_2010_chemical_composition). Finally, their enrichment factor (ratio of their average concentration in the volcanic ash and the Earth's crust) is > 2 (Schäfer et al., 2011). Ti, in particular, can be considered a source-specific tracer for volcanic aerosol, or more in general, for crustal materials, since its background values at CMN are very low (Marenco et al., 2006), as opposed to Fe, Al and Mn, which, in some cases, can be influenced by anthropogenic sources. During the observation period, Ti concentrations measured by PIXE were above detection limits only in the samples corresponding to the two volcanic plume transits (maximum 73 $ng\,m^{-3}$ on 18 May). For Ti concentrations measured by ED-XRF at CMN in summer, Marenco et al. (2006) report mean and 90th percentile equal to 30 and 58 ng m^{-3} respectively, with peaks attributed to Saharan dust transports.

For all such elements, background concentrations measured on 11 May (a day not affected by volcanic particles) were subtracted from those recorded during the volcanic plume transits. In this way, the amount of volcanic aerosol present at CMN was estimated, taking into account the relative abundances of the four elements in the ash collected near the volcano. The mass content of TiO_2 in the volcanic aerosol was 0.3 % on 18 May, which was slightly lower than that observed at the Swiss free tropospheric station JFJ by Bukowiecki et al. (2011) during the same episode (0.5 %).

The impact of the volcanic ashes on PM_{10} at CMN was estimated to be in the range from 7.3 to 9.5 µg m^{-3} (average 9 ± 1 µg m^{-3}), i.e. between 32 % and 41 % (average 39 ± 4 %) of the total reconstructed mass of PM_{10} (23 µg m^{-3}). This indicates that the volcanic ash contribution was superimposed on a background PM_{10} mass of about 14 µg m^{-3}, a value very close to the average PM_{10} mass of 16 µg m^{-3} measured by Marenco et al. (2006) at the same site during a 3 month summer campaign in 2004. In May, on the basis of CO, O_3 and BC measurements at CMN (Marinoni et al., 2008; Cristofanelli et al., 2013), it can be assumed that the local meteorology already presents features typical of the warm period, when the site is affected by emission sources located in northern Italy and continental Europe, transported by valley breezes or convective activity from the boundary layer, even during the night (Marinoni et al., 2008). The volcanic plume almost doubled the background aerosol concentration at CMN, though this contribution was smaller than that observed in other parts of Europe. A summary of the volcanic contributions calculated from chemical tracers or estimated by models is reported in Table 2 for the two episodes separately. The table denotes a high variability in such estimates, reflecting the uncertainty in the basic data (composition of volcanic ash and PM_{10} sample analysis).

Compared to the April episode, the estimates of the volcanic contribution in May denote an overall better coherence, with decreasing intensity at increasing distance from the source, despite the small number of observations. On this occasion volcanic contributions of up to 70 % were observed in Switzerland (Bukowiecki et al., 2011). A 60 % volcanic contribution is reported at a high altitude station in France (Puy de Dome, 1465 m a.s.l.). According to the authors (Hervo et al., 2012), the 33 % Flexpart estimate at Puy de Dome, reported in Table 2 for the same site, represents an underestimate, partly explained by the fact that the model does not simulate the formation of secondary aerosols. By contrast, the April episode denotes a higher degree of variability in the volcanic contributions calculated at different sites. This probably reflects the various degrees of mixing between volcanic plume and PBL air during the episode, again suggestive of the different transport dynamics of the two described events, with the simultaneous presence in April at different sites of contributions other than the volcanic plume, which are difficult to quantify.

During the April event a 19 % volcanic contribution was estimated at CMN. At an even longer distance, in Puglia, southern Italy, Perrone et al. (2012) report a maximum volcanic ash contribution of 19 % to total PM_{10}, derived from Flexpart simulations. To our knowledge no similar estimates exist for other sites south of the Alps, although observations of the plume are reported over Spain and Greece.

3.7 PM_{10} mass concentration in the Po Valley

PM_{10} concentrations recorded by the Air Pollution Monitoring Network of the Regional Agency for Environmental Protection in the urban area of Bologna and in the lowland site of San Pietro Capofiume (SPC) (40 km north-east of Bologna in the Po Valley) were analysed to evaluate whether the volcanic plume had an impact on air quality also within the densely populated Po Valley. Data did not evidence any marked increase in PM_{10} concentrations for the April episode, while a relative maximum was observed on 19 May, more pronounced for particle sizes larger than 2.5 µm (Fig. 8). The weak signal recorded by these low altitude stations is probably attributable to the strong dilution of the volcanic plume combined with the masking effect of anthropogenic pollution. In fact, both sites are characterized by much higher mass concentrations of particles, compared to the background site of CMN. Hence a relatively small volcanic contribution superimposed on a much higher background would be hard to distinguish.

Conversely, the correspondence of the relative peak of 26 April with the accumulation mode peak observed at CMN (Fig. 1) leads us to attribute this peak to pollution transport from the PBL, in the presence of a persistent high pressure period. Measurements of optical and physical aerosol properties performed at the same site of SPC by a ground-based radiometer (Campanelli et al., 2012) confirm that, at least during the April episode, the transit of the volcanic plume was not unequivocally detectable, due to the mixing with high particulate matter concentrations typical of the Po Valley.

4 Conclusions

The paper presents the results of continuous measurements of aerosol physical parameters and off-line chemical analyses of aerosol samples collected during April and May 2010. Such data allow the detection of the Eyjafjallajökull volcanic plume transit over CMN, after a long-range transport of more than 3000 km. During the eruption period, two main events impacted the site, the first on 19–21 April and the second between 18 May and 20. The two events were both identified by a sudden increase in the fine mode particle number, accompanied by a proportionally higher build-up of the coarse particle number. The presence of volcanic emissions in the free troposphere at CMN was confirmed by back-trajectory analysis and by the off-line analysis of the chemical composition of the aerosol, which exhibited chemical features typical of volcanic ash, starting with an increase in sulphate concentration.

The presented data show that, in both the episodes described here, the volcanic plume reached CMN strongly diluted as a result of the long distance from the source. The simultaneous increase in the concentrations of elements such as Fe, Al, Mn, Ti and Cr support the volcanic nature of the transported materials.

The ash (coarse fraction) was mainly composed of silicate glass and crystalline material, reflecting the chemical composition of the magma from which it derived and that of the conduit rocks eroded during the eruption, largely consisting of silica and aluminium oxides, with smaller proportions of other oxides, materials that do not easily solubilise in water. By contrast, the fine aerosol fraction during the two episodes appears to be mainly composed of acidic sulphates, produced by the oxidation of the SO_2 degassed by the volcano, which subsequently condensed on pre-existing or freshly nucleated particles.

The concentration of sulphate in PM_1 during the most intense event of 18 May reached a relative maximum not correlated with other pollutants, such as nitrate or organic carbon, a feature typical of air parcels of volcanic origin. Data of specific volcanic tracers measured at CMN by PIXE-PIGE i.e. Fe, Ti, Al, Mn, and the concentrations of the same elements in the ash collected in Iceland near the source, allowed the reconstruction of the PM10 mass contributed by the volcanic plume at CMN. The calculated contribution ranged from 7.3 to 9.5 $\mu g\,m^{-3}$, corresponding respectively to 32 to 41 % of the reconstructed mass of $PM_{10.}$

To our knowledge, no similar estimates were provided for other sites south of the Alps, though plume observations were reported in the literature for Spain, Slovenia and Greece.

The impact of the volcanic plume at CMN did not strongly affect the air quality at ground level. In fact, $PM_{2.5}$ and PM_{10} data of the Regional Agency for Environmental Protection in the urban area of Bologna and at the lowland site of San Pietro Capofiume did not evidence any significant increase in aerosol mass during the episodes. This emphasises the importance of aerosol chemical analysis in distinguishing volcanic influences from other factors and processes, and confirms that the high altitude site of CMN, due to its remote, mostly non-PBL-influenced location, is an appropriate observation site, not only for characterizing the regional background, but also for identifying air-mass transport effects and isolating long-range transport phenomena, such as the volcanic plume transport over southern Europe.

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Table 1. Average and peak values of particle concentrations (accumulation mode, coarse mode and total number), scattering and absorption coefficient, concentration of major inorganic ions and water soluble organic carbon (WSOC) at CMN during the two volcanic events and during a 5 yr reference period. Accumulation and coarse particle concentration are averaged on a 30 min basis, particle total number concentration and scattering/absorption coefficients on a 60 min time basis. [a]The reference period for the particle total number concentration encompasses the years 2008–2010. [b]The reference period for sulphate, nitrate and ammonium encompasses years 2009–2011.

	Event I 19–21 Apr 2010	Event II 18–20 May 2010	Apr–May 2006–2011
Accumulation mode particles (cm^{-3})	60.8 ± 4.4; Max: 122.8	41.1 ± 7.2; Max: 245.0	21.8 ± 28.5; Max: 245.0
Coarse particles (cm^{-3})	0.57 ± 0.04; Max: 1.3	1.04 ± 0.18; Max: 4.3	0.20 ± 0.43; Max: 4.9
Total particle number (cm^{-3})	2026 ± 173; Max: 4544	4102 ± 562; Max: 6831	2486 ± 165; Max: 10469[a]
Scattering coeff. (Mm^{-1})	47.5 ± 4.1; Max: 84.9	39.9 ± 6.4; Max: 134.5	22.2 ± 20.5; Max: 163.0
Absorption coeff. (Mm^{-1})	3.37 ± 0.28; Max: 5.99	2.39 ± 0.28; Max: 5.54	1.60 ± 1.78; Max: 18.74
SO$_4$, PM$_1$/PM$_{1-10}$ (µg m^{-3})	2.14/0.22	3.35/1.69	0.41 ± 0.90/0.10 ± 0.13[b]
NO$_3$, PM$_1$/PM$_{1-10}$ (µg m^{-3})	0.93/0.31	0.43/0.16	0.24 ± 0.47/0.17 ± 0.38[b]
NH$_4$, PM$_1$/PM$_{1-10}$ (µg m^{-3})	0.49/ < DL	1.10/0.01	0.15 ± 0.37/0.005 ± 0.008[b]

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Table 2. Summary of volcanic contributions to total PM$_{10}$ as observed at different sites in Europe and estimated by the indicated method.

Author	Site	Alt. (m)	PM$_{10}$ (µg m^{-3})	Volcanic contrib. (µg m^{-3})	% on PM$_{10}$	Method
Episode 19 Apr–21						
Pitz et al. (2011)	Augsburg – Germany (48°22′ N 10°54′ E)	484		12 (av.) 35 (max)	30 (av.) 65 (max)	Positive Matrix Factorization
Schäfer et al. (2011)	Augsburg – Germany (48°22′ N 10°54′ E)	484		17 ± 7	25 ± 10	Chemical tracers (Ti, Mn, Sr, Y, Zr)
	Andechs – Germany (47°58′ N 11°11′ E)	690		10 ± 4	26 ± 10	Chemical tracers
Colette et al. (2011)	Mulhouse – France (47°45′ N 7°19′ E)	240	56	30 ± 10	54 ± 18	Chemical tracers (Al, Fe, Ti)
Bukowiecki et al. (2011)	Basel – Switzerland (47°34′ N 7°36′ E)	260	51.5	45	87	Chemical tracers (Ti)
Perrone et al. (2012)	Lecce – Italy (40°21′ N, 18°10′ E)	50	24	4.5	19	Flexpart simulation
This study	**Mt. Cimone – Italy (44°11′ N, 10°42′ E)**	**2165**	18 (reconstr.)	**3 ± 2**	**18 ± 9**	**Chemical tracers (Al, Fe, Ti, Mn)**
Episode 18 May–20						
Bukowiecki et al. (2011)	Lugano – Switzerland (46°00′ N, 8°57′ E)	280	26.1 (24 h)	18.8	72	Chemical tracers (Ti)
	Magadino – Switzerland (46°08′ N 8°51′ E)	204	26.3 (24 h)	18.3	70	Chemical tracers (Ti)
Hervo et al. (2012)	Puy de Dome – France (45°46′ 0 N, 2°57′ E)	1465	39 ± 4 (TSP)	13 ± 4	30	Flexpart simulation
			52 ± 9	30 ± 4	60	PM$_{10}$ difference from background conc.
This study	**Mt. Cimone – Italy (44°11′ N, 10°42′ E)**	**2165**	**23 (reconstr.)**	**9 ± 1**	**37 ± 5**	**Chemical tracers (Al, Fe, Ti, Mn)**

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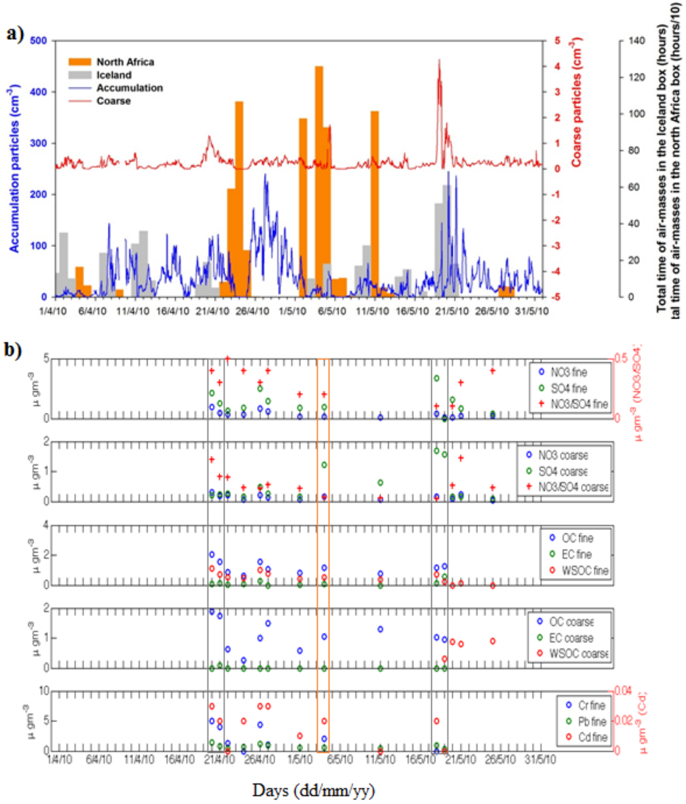

Fig. 1. (a) Particle number concentration recorded at CMN from 1 April to 31 May 2010 measured by OPC (0.3–20 µm), separately for the accumulation mode (< 1 µm) and coarse mode (> 1 µm). The vertical bars denote the daily total number of back-trajectory points falling within the "Iceland" (grey) and the north Africa (orange) geographical boxes. **(b)** Temporal evolution of sulphate, nitrate, NO_3/SO_4 ratio, OC, EC and WSOC, separately for the fine and coarse mode, and Cr, Pb and Cd in the fine fraction.

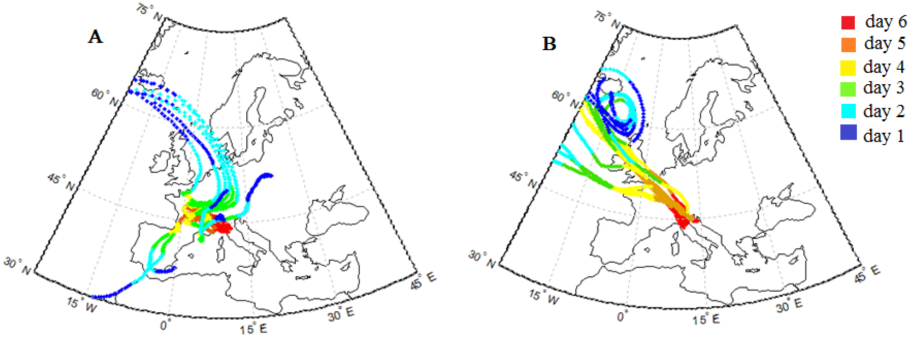

Fig. 2. Member of the back-trajectory ensembles travelling over the "Iceland" box for the two "major" events 19–21 April **(A)** and 18–20 May 2010 **(B)**. The different colours identify the day of transport, starting from the source at day 1 and ending at CMN at day 6.

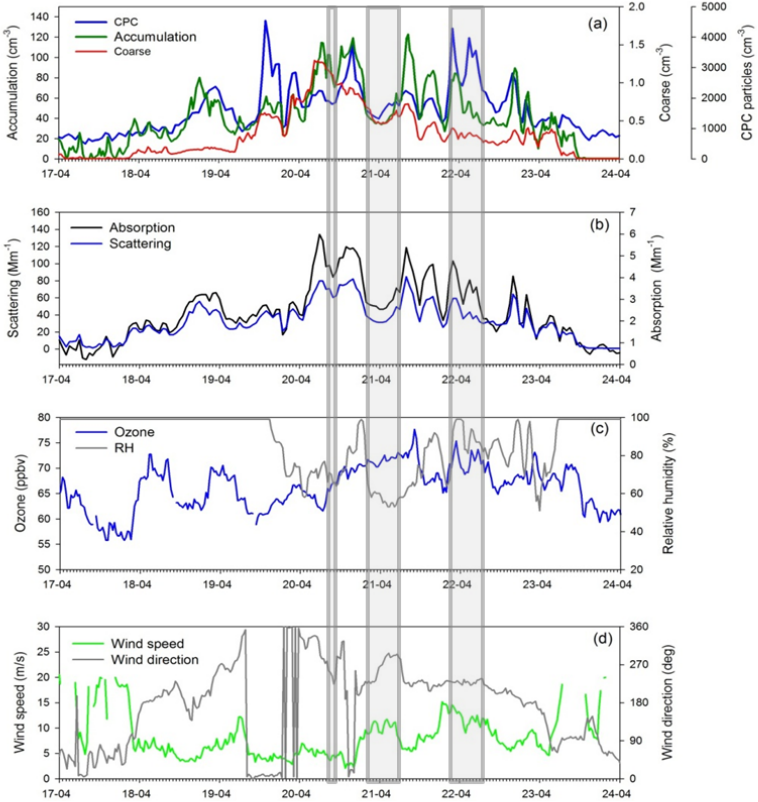

Fig. 3. April 2010: time series of **(a)** particle concentration (cm^{-3}) in the CPC, accumulation and coarse mode, **(b)** absorption and scattering coefficient, **(c)** surface ozone and relative humidity and **(d)** wind speed and wind direction. The time periods covered by samples for chemical analyses are shaded.

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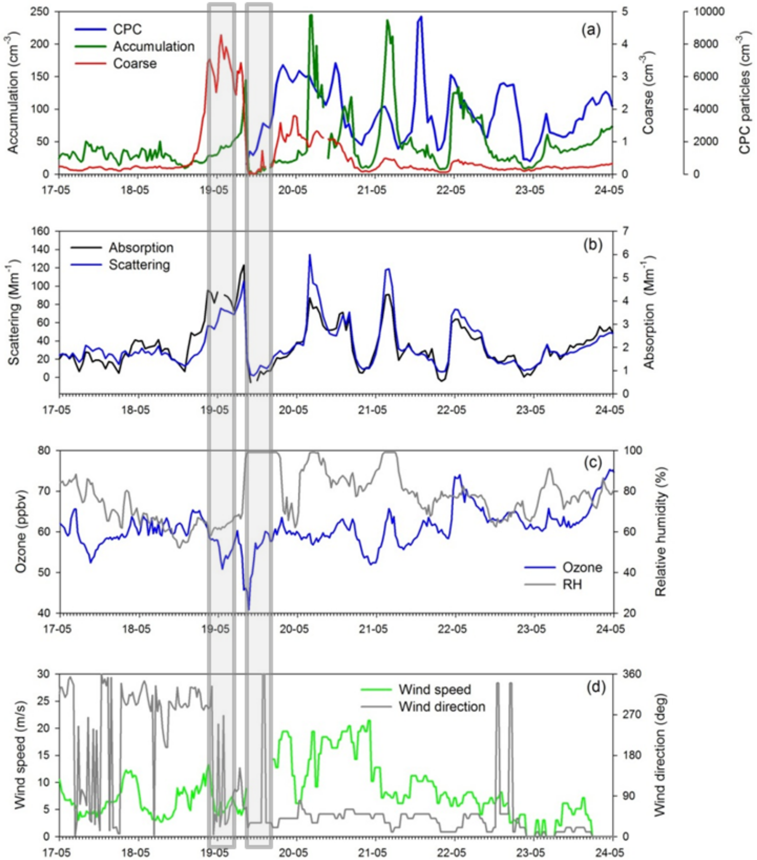

Fig. 4. May 2010: time series of **(a)** particle concentration (cm^{-3}) in the CPC, accumulation and coarse mode, **(b)** absorption and scattering coefficient, **(c)** surface ozone and relative humidity and **(d)** wind speed and wind direction. The time periods covered by samples for chemical analyses are shaded.

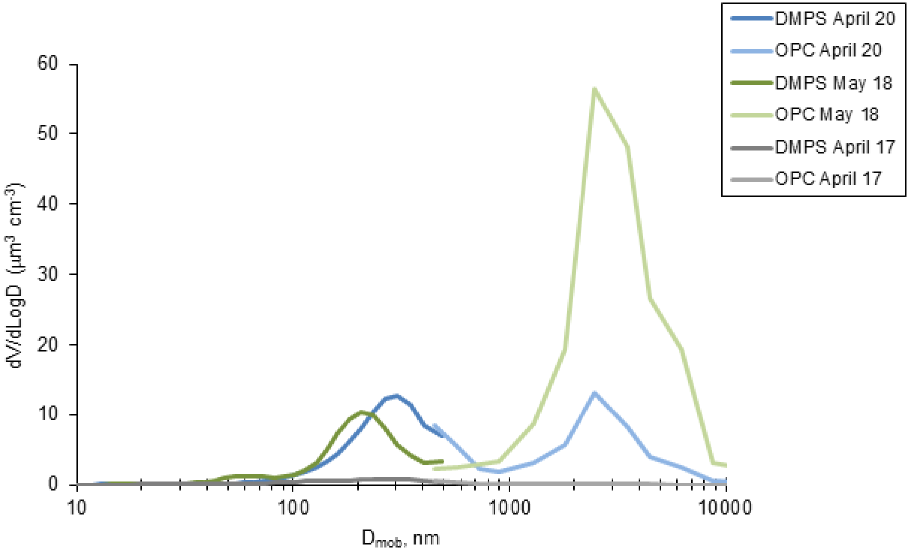

Fig. 5. Comparison between the aerosol size distribution before the arrival of the volcanic plume, 17 April (black/grey) and during its most intense impact on 20 April (blue/light blue) and 18 May (green/light green), derived from DMPS (10–500 nm) and OPC (300 nm–20 µm) measurements.

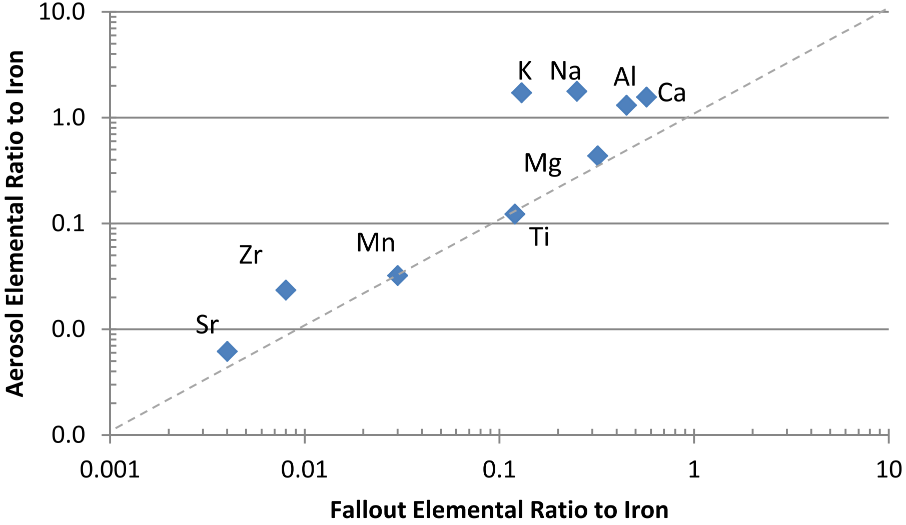

Fig. 6. Elemental ratio to iron in the aerosol sample collected at CMN on 18 May 2010 vs. direct fallout collected on 16 May (GSV165-3) near the Eyjafjallajokull crater (http://earthice.hi.is/eyjafjallajokull_2010_chemical_composition). The dashed line indicates equal ratio.

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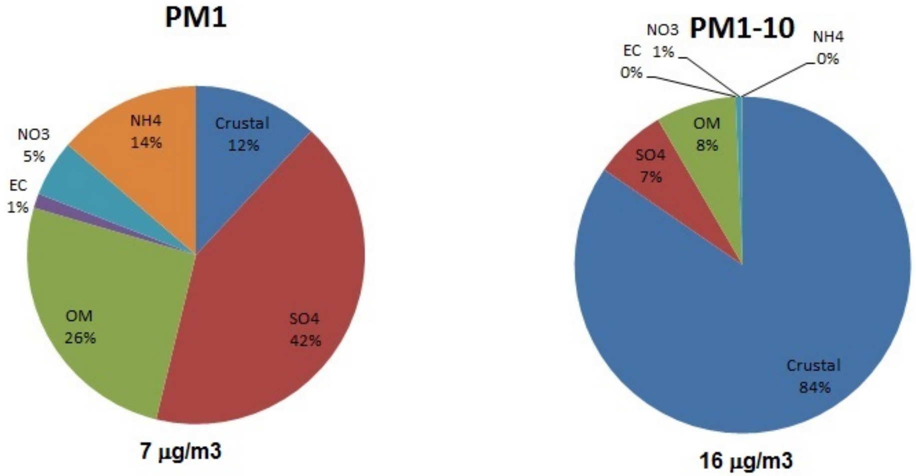

Fig. 7. Reconstructed mass for the 18 May episode, separated into the fine (left) and coarse (right) fraction. The reconstructed mass is indicated below the figure. The total PM_{10} reconstructed mass was 23 $\mu g\,m^{-3}$.

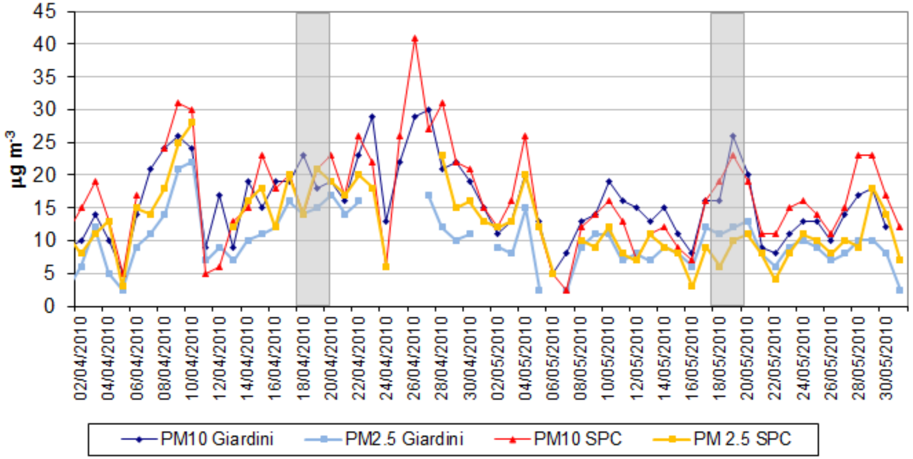

Fig. 8. PM_{10} data of the Regional Agency for Environmental Protection collected at the Bologna urban background station Giardini Margherita and at the rural background station of San Pietro Capofiume (SPC) (http://service.arpa.emr.it/qualita-aria/bollettino.aspx?prov=bo).

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