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Particle size distribution factor as an indicator for the impact of the Eyjafjallajökull ash plume at ground level in Augsburg, Germany

M. Pitz^{1,2}, J. Gu^{1,2}, J. Soentgen², A. Peters¹, and J. Cyrys^{1,2}

¹Helmholtz Zentrum München, German Research Center for Environmental Health, Institute of Epidemiology II, 85764 Neuherberg, Germany

²University of Augsburg, Environment Science Center, 86159 Augsburg, Germany

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Correspondence to: M. Pitz (mike.pitz@hs-augsburg.de)

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Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

During the time period of the Eyjafjallajökull volcano eruption in 2010 increased mass concentration of PM₁₀ (particulate matter, diameter <10 µm) were observed at ground level in Augsburg, Germany. In particular on 19 and 20 April 2010 the daily PM₁₀ limit value of 50 µg m⁻³ was exceeded. Because ambient particles are in general a complex mixture originating from different sources, a source apportionment method (positive matrix factorization; PMF) was applied to particle size distribution data in the size range from 3 nm to 10 µm to identify and estimate the volcanic ash contribution to the overall PM₁₀ load in the ambient air in Augsburg.

A PMF factor with relevant particle mass concentration in the size range between 1 and 4 µm (maximum at 2 µm) was associated with long range transported dust. This factor increased from background concentration to high levels simultaneously with the arrival of the volcanic ash plume in the planetary boundary layer. Hence, we assume that this factor could be used as an indicator for the impact of the Eyjafjallajökull ash plume on ground level in Augsburg.

From 17 to 22 April 2010 long range transported dust factor contributed on average 30.2 % (11.6 µg m⁻³) to PM₁₀. On 19 April 2010 at 20:00 UTC+1 the maximum percentage of the long range transported dust factor accounted for around 65 % (35 µg m⁻³) to PM₁₀ and three hours later the maximum absolute value with around 48 µg m⁻³ (61 %) was observed. Additional PMF analyses for a Saharan dust event occurred in May and June 2008 suggest, that the long range transported dust factor could also be used as an indicator for Saharan dust events.

1 Introduction

Particulate matter (PM) is ubiquitously present in the atmosphere and originate from natural processes (e.g. erosion (soil, sea salt, desert dust), forest and bush fires, volcanoes) and anthropogenic emissions (e.g. traffic, industry, domestic heating, agriculture)

ACPD

11, 16417–16437, 2011

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



as well as from formation in the atmosphere by nucleation processes or from gaseous precursors (secondary aerosol formation). Hence, the chemical composition of ambient PM is very complex and variable and finally, the composition of particles affects their transport and residence time in the atmosphere, solar radiation and deposition in the environment. All these factors in turn affect ambient air quality. For example, Birmili et al. (2008) showed that a wind-blown soil dust event in southern Ukraine led to extremely high mass concentration of PM₁₀ (aerodynamic diameters <10 µm) in central Europe.

In the European Union limit values for the mass concentration of PM₁₀ were established since 2005 “to avoid, prevent or reduce harmful effects on human health and the environment as a whole” (Council Directive 1999/30/EC). The Directive 2008/50/EC permits under specific conditions (“... determined with sufficient certainty, and where exceedances are due in whole or in part to these natural contributions...”) the subtraction of (I) re-suspension of particles (e.g. winter-sanding or -salting of roads) or (II) of natural source contributions (e.g. volcanic ash, desert and soil dust) from the measured PM mass concentration. The effects of natural sources on PM ground level are difficult to prove. First of all supplemental analyses of the origin of the air masses are mostly needed (e.g. satellite images, backward trajectories). In addition to PM₁₀ mass concentration normally further measurements such as elemental composition and gaseous parameters are necessary to estimate the contribution of the natural sources to the PM burden.

In this study, we will demonstrate a method for relatively rapid identification of natural dust events and estimation of their contribution to the increase of PM₁₀ mass concentration in Augsburg, Germany using as an example the eruption of the Eyjafjallajökull volcano in Iceland, which was visually observed for the first time on 20 March 2010 (IES, 2010). The trajectory of the volcanic ash plume was tracked by the London Volcanic Ash Advisory Centers (VAAC) using a combination of infrared channels of satellite images and atmospheric dispersion modeling (VAAC, 2010). As a result of the advection of volcanic ash clouds over Europe, no-flying zones were established to

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



prevent damage on aircrafts, which led to a closure of the most European airports from 15 to 21 April 2010.

The presence of the volcanic ash plume over Europe has been also demonstrated by aircraft measurements conducted directly in the Eyjafjallajökull ash plume e.g. by the research aircraft Falcon from the German Aerospace Center (Schumann et al., 2011). Some other studies demonstrate the presence of the volcanic ash plume indirectly by ground-based remote sensing measurements (e.g. Colette et al., 2011; Flentje et al., 2010; Gasteiger et al., 2011; Schäfer et al., 2011). Emeis et al. (2011) described the temporal and spatial structure of the volcanic ash plume by numerical simulations with the Eulerian meso-scale model. Thomas and Prata (2011) studied the feasibility of using sulphur dioxide (SO_2) as a tracer for the volcanic ash plume by comparing ash retrievals from the geosynchronous Meteosat satellite with SO_2 measurements based on satellite sensors received from polar orbiting platforms.

Flentje et al. (2010) observed enhanced SO_2 and particle number concentration levels simultaneously with the arrival of the Eyjafjallajökull ash plume in the time period 16 to 21 April 2010 at two mountain stations in the northern Alpine region in Germany; the Environmental Research Station (UFS) "Schneefernerhaus" (located 2650 m a.s.l., 300 m below the Zugspitze summit) and the Hohenpeissenberg Meteorological Observatory HPB (985 m a.s.l., 300 m above the surrounding countryside). However, the volcanic impact at HPB was partially masked by enhanced anthropogenic background. Colette et al. (2011) reported the most outstanding enhanced levels of PM_{10} mass concentration on 18 and 19 April 2010 for an urban background station in northern France (Mulhouse). A combination of atmospheric dispersion modelling, ground-based remote sensing measurements and chemical analyses of sampled particles were used for the estimation of the volcanic ash impact to the observed increase of ambient PM_{10} mass concentration. Schäfer et al. (2011) observed elevated particle number concentrations of ultrafine particles (UFP, diameter $<100\text{ nm}$) for the regional background stations UFS, HPB, and SSL ("Schauinsland", 1210 m a.s.l.) which can be assigned to particle formation in the volcanic ash plume from the gaseous precursor SO_2 . In contrast,

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



increased UFP at an urban background site in Augsburg, Germany could not be clearly identified as of volcanic origin because of the higher anthropogenic impact. The identification and quantification of volcanic ash impact to ambient PM_{10} levels measured on 19 and 20 April 2010 at five monitoring stations of the Bavarian air monitoring network (Andechs and Tiefenbach: regional background, Augsburg: urban background, München and Oberaudorf: traffic) was conducted in the Schäfer study by means of additional measurements, e.g. titanium content in PM_{10} . Moreover, Schäfer et al. (2011) observed an increase of sulphate mass concentration in $PM_{2.5}$ without any major SO_2 peak during the volcanic ash plume episode in Augsburg. It indicates that a significant amount of the initial emitted SO_2 was converted to sulphuric acid (H_2SO_4) and sulphate mass concentration during transport of the volcanic ash plume. The SO_2 concentrations in the volcanic ash plume were additionally reduced by increased dilution during the transport down to the monitoring site in Augsburg (484 m a.s.l.) and good vertical mixing and advection conditions. This assumption is supported by the observation that despite of rather low SO_2 levels in Augsburg, an increase of SO_2 concentrations was observed at monitoring sites at higher elevation, such as UFS or HPB site.

It indicates, that the identification of such natural events as volcanic ash plume, which could lead to a dramatically increase of ambient particle concentrations, is more difficult in urban areas than in rural or mountain areas. The reason for this is the impact of anthropogenic background which is much stronger in urban areas in comparison to rural areas or even mountain stations.

In this study we applied positive matrix factorization method using particle size distribution data in the diameter range 3 nm to 10 μm for identification of the Eyjafjallajökull ash plume and estimation of the contribution of the Eyjafjallajökull ash plume to the PM_{10} mass concentration in Augsburg, Germany.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2 Experimental

2.1 Sampling location

The measurement site is located on the premises of the University of Applied Sciences (AUHS, 48°22' N; 10°54' E; 484 m a.s.l.) about 1 km south of the city center and is representative for urban background conditions in Augsburg, Germany (Cyrus et al., 2008). Additional details of the site have been reported by Pitz et al. (2008a, b). The volcano Eyjafjallajökull, Iceland is located around 2500 km away from Augsburg, Germany.

Because no gaseous pollutants were measured at the AUHS site, SO₂ concentration from a measurement site of the Bavarian air monitoring network was used. This site at the Bavarian State Office for the Environment (AUBÜ) is located about 3.5 km away south of the AUHS site and is also considered as an urban background site.

2.2 Measurement methods

A Twin Differential Mobility Particle Sizer (TDMPs) system combined with an Aerodynamic Particle Sizer (APS, model 3321, TSI Inc., USA) was used to measure particle size distribution in the diameter range 3 nm to 10 µm. Additional information about the merging of TDMPs and APS data as well as for the evaluation of the calculated mass concentration from the merged size distribution can be found in the appendix A. The Black Carbon (BC) fraction of PM_{2.5} was measured with an Aethalometer (series 8100, Thermo Fisher Scientific Inc., USA). Details of the measurement devices and the applied quality assurance have been reported by Pitz et al. (2008a, b) and Birmili et al. (2010). Briefly, the merged particle size distribution and all relevant parameters of the devices were visually checked normally on daily base. In addition, for the APS (measurement range >0.8 µm) a zero-point check and a check with monodisperse latex particles (1.06 ± 0.04 µm) was performed on monthly base.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.3 Positive matrix factorization (PMF)

We used the receptor model PMF 3.0 from the US Environmental Protection Agency (EPA) to identify particle size distribution factor profiles and quantify factor contributions. PMF is a widely used receptor model for characterizing aerosol sources (Paatero, 1999). It decomposes the measured PM composition data (or size distribution data etc.) into sub factor profiles and factor contributions. Comparing it with previously used methods like principal components analysis, it has the advantage of more realistic non-negative constraints on factor profiles and contributions, and better scaling of the data by individually assigned uncertainties (Paatero and Tapper, 1994; Paatero, 1997). The application of PMF to particle size distribution data is in parts more difficult to interpret because specific tracers are not present as normally available for chemical composition data. Hence, for an interpretation of the PMF factors obtained from particle size distribution data additional data are necessary.

Because of marginal data coverage, three size channels (3 nm, 3.4 nm and 10 μ m) were excluded from the analyses, finally resulting in 64 size channels (3.8 nm to 8.8 μ m). Four channels between 3.8 and 5.5 nm, four channels between 0.75 and 1 μ m and three channels between 6.9 and 8.8 μ m were set to weak.

The measurement of particle size distribution provides no detection limit; hence we assumed the uncertainty of every channel to 10 % (Yue et al., 2008). If the number concentration of a channel was zero, we assumed an uncertainty of 10 % of the total mean of the respective channel. Moreover, an extra modeling uncertainty of 25 % was applied to every size channel. The PMF analyses were performed for the time period 1 April to 31 May 2010 (Eyjafjallajökull volcano eruption) and in addition for the time period 1 May to 30 June 2008 (Saharan dust event).

Initially, we analyzed and compared different numbers of factors and we compared the results subsequently with each other. Based on these preliminary investigations PMF model with seven factors reveal the most plausible and interpretable results to characterize the potential particle sources. Moreover, multiple bootstrap runs were performed to test the stability and uncertainty of the model results.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



PMF results observed in this study showed the same number and profiles of the particle size distribution factors as already shown by Gu et al. (2011) for the same site in Augsburg in winter 2006/2007 where additional gaseous pollutants, online chemical compositions and meteorological data were used to assist interpreting the PMF results.

3 Results and discussion

In Fig. 1 the time series of the hourly average PM₁₀ mass concentration measured at the University of Applied Sciences (AUHS) and the SO₂ mass concentration measured at the Bavarian State Office for the Environment (AUBÜ) in the time period 4 April to 4 May 2010 is shown for Augsburg, Germany. In particular on 9 April and 19/20 April 2010 increased PM₁₀ mass concentrations could be observed whereas the daily PM₁₀ limit of 50 µg m⁻³ was only exceeded on 09 April at the AUHS site. At the other four measurement sites of the Bavarian air monitoring network in Augsburg partially exceeding of the respective threshold during the two time periods could be observed (data not shown). On the other hand, no increased SO₂ concentrations were observed in the same time periods at ground level in Augsburg which is in contrast to other monitoring sites where elevated SO₂ levels were observed in parallel to elevated PM₁₀ levels (Flentje et al., 2010; Schäfer et al., 2011). Therefore, the clearly identification of the volcanic ash impact on ground level in Augsburg was not possible without additional information. In other words, it was not possible to distinguish whether the increased PM₁₀ levels in Augsburg were caused by the Eyjafjallajökull ash plume or if the observed increased PM₁₀ levels were caused by other local anthropogenic sources. Hence, PMF analysis was applied to particle size distribution data to identify the possible sources. The performed PMF analyses for April to May 2010 resulted in seven source factors with similar factor profiles as already found by Gu et al. (2011) for Augsburg in winter 2006/2007. As described by Gu and colleagues those factors could be associated to (I) nucleation particles, (II) fresh traffic emissions, (III) aged traffic emissions, (IV) stationary combustion, (V) secondary aerosols, (VI) re-suspended dust, and

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(VII) long range transported dust. For more details regarding the interpretation of the PMF factors please refer to Gu et al. (2011).

In Fig. 2 the time series section from 4 April to 4 May 2010 for the normalized factor contribution to PM_{10} (the average of all contributions for each factor is 1 for the total time period April to May 2010) and the percentage of particle mass size distribution of the factor which was associated to long range transported dust is shown. The relevant mass concentration size range for the long range transported dust factor was between 1 and 4 μm with the peak maximum at 2 μm . A remarkable large peak of the long range transported dust factor was found around midnight on 19 April which coincided with the PM_{10} peak shown in Fig. 1. It seems that the long range transported dust factor was in fact (at least partially) influenced by the Eyjafjallajökull ash plume. Our findings for increased PM_{10} mass concentration in parallel to the increase of the factor contribution agree very well with the observed findings by Schäfer et al. (2011) at the same site. In the Schäfer study increased secondary sulphate mass concentrations during the volcanic ash impact were observed which were attributed to the formation from the gaseous precursor SO_2 during transport of the Eyjafjallajökull ash plume. Moreover, our findings are in good agreement with the findings of Schumann et al. (2011). During the flight with the research aircraft Falcon over southern Germany minor ash mass concentrations on 9 April 2010 and increased ash mass concentration on 19 April were observed.

Figure 3 shows the hourly average particle volume size distributions (spherical shape assumed) for the time period largely affected by the volcanic ash plume (19 April 19:00 to 20 April 10:00 UTC+1) in comparison to the hourly average of the long-term time period 2005 to 2010. Normally, a bimodal particle size distribution with a prevalent bias to particles smaller than 1 μm could be observed as indicated by the blue line representing the long-term hourly average. During the volcanic ash plume impact episode a drastic increase in particular of particles with diameters greater than 1 μm could be observed.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



As shown in Fig. 4 starting on 17 April lasting for about 6 days until 22 April 2010 increased mass concentrations of the long range transported dust factor could be observed. In this time period the long range transported dust factor contributed on average 30.2 % ($11.6 \mu\text{g m}^{-3}$) to the PM_{10} mass concentration. In the evening of 19 April 2010 at 20:00 UTC+1 the maximum percentage of the Eyjafjallajökull ash plume to the PM_{10} mass concentration with around 65 % ($35 \mu\text{g m}^{-3}$) was observed. Three hours later the maximum absolute value accounted for around $48 \mu\text{g m}^{-3}$ (61 %) to the PM_{10} mass concentration. During the non-affected time period from April to May the long range transported dust factor contributed on average 14 % ($2.5 \mu\text{g m}^{-3}$) to the PM_{10} mass concentration. Additional analyses of the BC concentration at the AUHS site showed weak correlation between the long range transported dust factor and the BC mass concentration. Nevertheless, on 20 April at 06:00 UTC+1 in the morning increased BC mass concentrations were observed in parallel to increased mass concentrations of the long range transported dust factor and PM_{10} (Fig. 4). It means, that the second PM_{10} mass concentration peak ($82.8 \mu\text{g m}^{-3}$) was associated not only to volcanic ash plume ($32.0 \mu\text{g m}^{-3}$), but also to local particles generated by anthropogenic sources in the city of Augsburg which could be mainly assigned to stationary combustion ($17.4 \mu\text{g m}^{-3}$), fresh traffic ($7.2 \mu\text{g m}^{-3}$) and aged traffic ($3.4 \mu\text{g m}^{-3}$) emissions.

In the recent past Bruckmann et al. (2008) described an outbreak of Saharan dust between 27 May and 1 June 2008 causing high PM_{10} levels north of the Alps. To test whether the contribution of the long range transported dust factor to PM_{10} levels is also increased during the Saharan dust outbreak (and not only during the volcanic ash episode), we also applied PMF method for the time period May to June 2008. PMF analyses resulted also in seven particle size distribution source factors with similar factor profiles as already above-mentioned for the time period April to May 2010 where the Eyjafjallajökull ash plume impact was observed. In Fig. 5 the time series section of the PM_{10} mass concentration and the normalized PM_{10} contribution of the long range transported dust factor from 4 April to 4 May 2010 for the Eyjafjallajökull ash plume

impact (Fig. 5a) and from 14 May to 31 June 2008 for the Saharan dust event (Fig. 5b) are comparative shown for the monitoring site at the University of Applied Sciences in Augsburg. A clear increase of the long range transported dust factor contribution was also observed during the Saharan dust event which correlates with the increased PM₁₀ levels. It indicates that the long range transported dust factor could be used for identification of such natural events as volcanic ash plume in April 2010 as well as Saharan dust episodes. A check of the contribution of the long range transported dust factor to PM₁₀ of the years 2005 to 2010 revealed that such events occur approximately once to twice per year.

4 Conclusions

PM₁₀ and SO₂ concentration which are normally available in air monitoring networks could not be used for identification and quantification of natural dust impact as the Eyjafjallajökull ash plume at urban ground level in Augsburg, Germany without any additional data. Normally, additional analyses of daily PM₁₀ filter samples for chemical composition are necessary to estimate the volcanic ash plume contribution to the PM₁₀ levels (assuming that the chemical composition of the volcanic ash is known). In this study, a PMF factor which was associated to long range transported dust (mass peak between 1 and 4 µm with a maximum at 2 µm) was used as an indicator for the impact of the Eyjafjallajökull ash plume at ground level in Augsburg. On 19 April 2010 at 20:00 UTC+1 the maximum contribution of the long range transported dust factor to PM₁₀ with about 65 % (35 µg m⁻³) was observed and the maximum absolute value of around 48 µg m⁻³ (61 %) was observed three hours later. Moreover, it could be shown that the long range transported dust factor could also be used as an indicator for a Saharan dust event in May and June 2008.

We showed in our study, that source apportionment using PMF method applied to hourly average ambient particle size distribution data offers a quite simple possibility to separate sources of a heterogeneous ambient mixture of different particulate sources.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



For the identification of natural sources (e.g. volcanic ash or desert dust), which occur for example in Augsburg approximately once to twice per year considering 2005 to 2010, particle size distribution (also of super micron particles) and additional information of the origin of the air masses such as satellite images or backward trajectories should be available.

Appendix A

Apparent particle density

For the conversion of the particle size distribution to mass concentration a spherical shape of the particles and an apparent mean density have to be assumed. Moreover, an effective density for the adaption of the aerodynamic diameter of the APS and the mobility diameter of the TDMPs has to be assumed.

In this study an effective density of 1.7 g cm^{-3} in the overlap range of 800–900 nm was assumed resulting in an excellent agreement of the merged TDMPs and APS size distributions as already shown by Pitz et al. (2008b).

For the calculation of the PM_{10} mass concentration from the merged size distribution of TDMPs and APS (3 nm to $10 \mu\text{m}$) a mean apparent density of 1.5 g cm^{-3} was used according to Pitz et al. (2003, 2008a, b). As demonstrated by Pitz et al. (2003, 2008a) the apparent particle density (calculated from PM mass concentration and particle volume concentrations measured in parallel by independent instruments) ranges from 1.0 to 2.5 g cm^{-3} for the 5th and 95th percentile, respectively depending on the particle composition.

The density of the directly emitted Eyjafjallajökull single ash particles vary between 0.7 and 3.2 g cm^{-3} (Schumann et al., 2011). The calculated densities for single particles sampled directly in the Eyjafjallajökull ash plume over the North Atlantic with the aircraft Falcon vary between 1.7 – 1.8 g cm^{-3} and 2.0 – 2.2 g cm^{-3} for particles $<0.5 \mu\text{m}$ and particles $>0.5 \mu\text{m}$ in diameter, respectively (Schumann et al., 2011). This raises

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the question whether the use of the mean apparent particle density for the whole study period (without specific density assumption for the volcanic dust events) led to additional uncertainties by the calculation of particle mass concentration from particle size distribution.

5 To answer the question, we calculated the average particle density for $PM_{2.5}$ and PM_{10} for the volcanic dust episode in Augsburg. No increase of apparent particle density could be observed, in contrast the apparent particle density for PM_{10} was 1.28 g cm^{-3} for the time period of the maximal volcanic ash impact on PM_{10} mass concentrations (19 April 19:00 to 20 April 10:00 UTC+1). It means that the average
 10 apparent particle density of PM_{10} was not changed by volcanic ash particles, probably due to contrary influence of other particulate components in the heterogeneous ambient mixture of different sources. Comparisons of hourly average calculated PM_{10} mass concentration (using the above-mentioned assumptions) from the size distribution of the TDMPS/APS system and independent measurements of PM_{10} by a Tapered
 15 Element Oscillating Microbalance (TEOM) in combination with an Filter Dynamics Measurement System (FDMS) at the same site from 17 to 22 April 2010 (the most important assumable impact of the volcanic ash plume on ground level in Augsburg) resulting in a slope of 0.96 and an intercept of $5.7 \mu\text{g m}^{-3}$ ($R^2 = 0.84$). Thus, the estimation of particle mass concentration (calculated from particle size distribution) for the volcanic
 20 ash event in Augsburg was not biased and it doesn't require any additional assumption regarding the apparent particle density.

As shown in Fig. 3 the volcanic ash particles contribute to particle mass concentrations mostly in the range between 1 and $10 \mu\text{m}$ in Augsburg. Assuming a mean particle density in this size range of 2.0 g cm^{-3} , we estimated the mean difference of
 25 PM_{10} mass concentration for the time period 17 to 22 April 2010 as “worst case” estimation. The calculated average underestimation is 10 % (minimum: 4 %, maximum: 15 %) compared to the PM_{10} mass concentration calculated by use of a mean apparent particle density of 1.5 g cm^{-3} for the entire size distribution.

Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Particle size distribution factor as an indicator

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Particle size
distribution factor as
an indicator**

M. Pitz et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

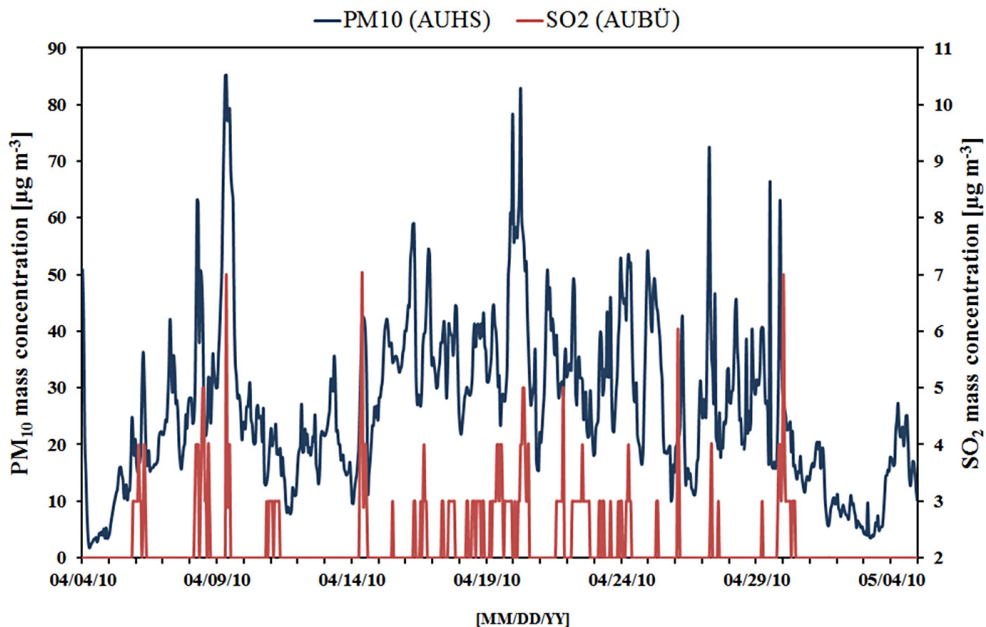


Fig. 1. Time series of hourly average PM_{10} mass concentration measured at the University of Applied Sciences (AUHS) and SO_2 mass concentration measured at the Bavarian State Office for the Environment (AUBÜ) in Augsburg, Germany.

**Particle size
distribution factor as
an indicator**

M. Pitz et al.

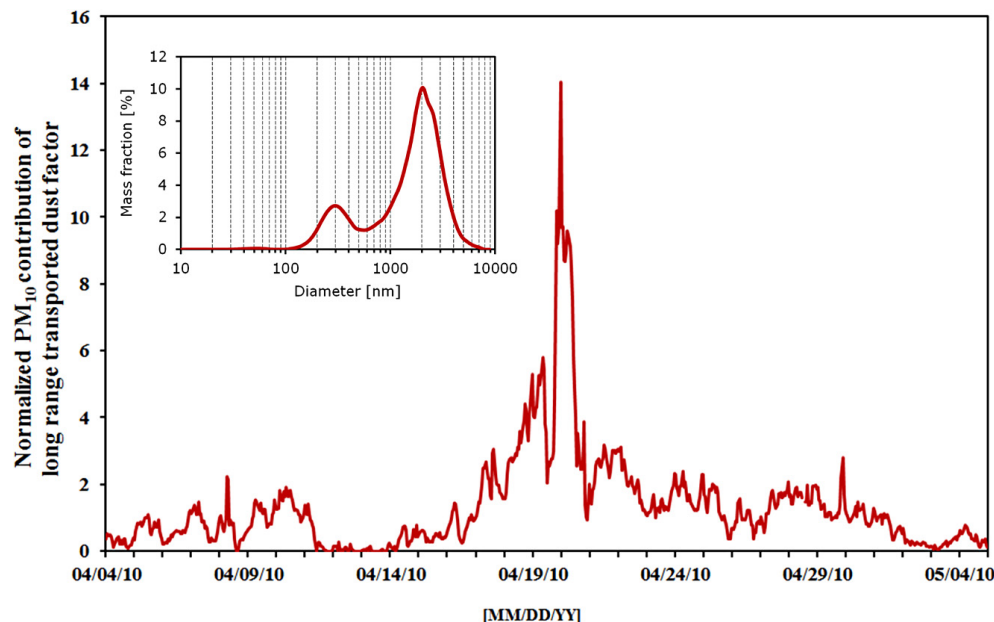


Fig. 2. Time series section of normalized hourly average PM_{10} contribution of long range transported dust factor at the University of Applied Sciences in Augsburg, Germany (average of all contributions for each factor is 1 for the total time period April to May 2010). The percentage of the particle mass size distribution is shown in the inset.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Particle size
distribution factor as
an indicator**

M. Pitz et al.

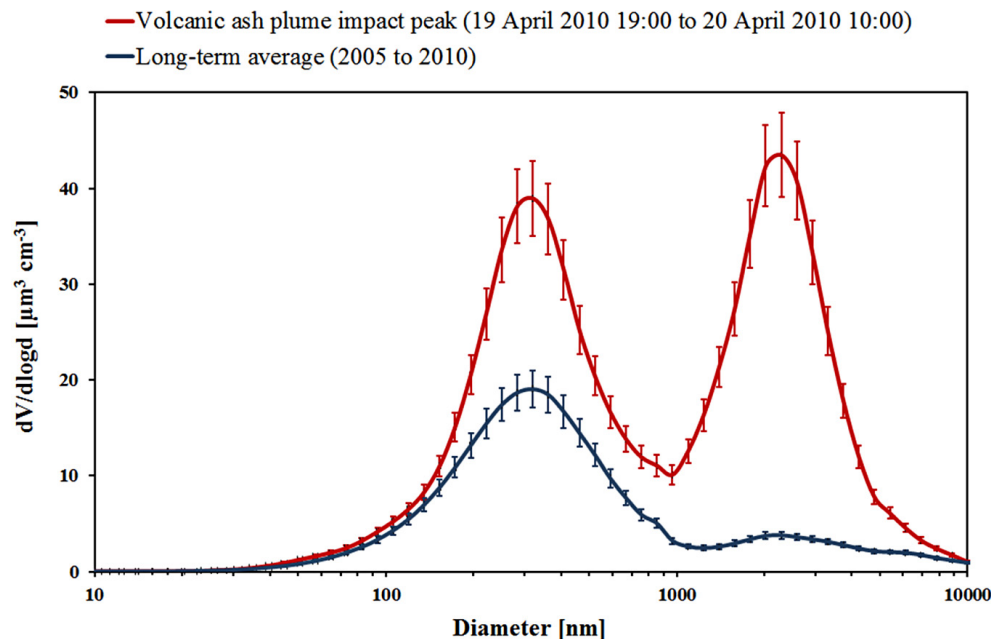


Fig. 3. Hourly average particle volume size distributions during the volcanic ash plume impact in comparison to the long-term average of the years 2005 to 2010 at the University of Applied Sciences in Augsburg, Germany. The error bars represent $\pm 10\%$.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Particle size distribution factor as an indicator

M. Pitz et al.

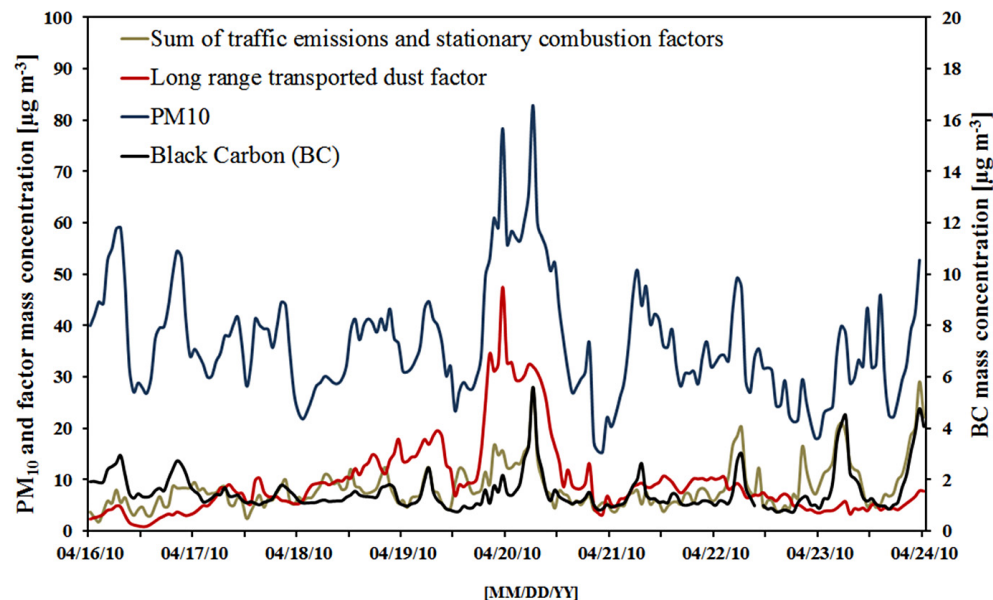


Fig. 4. Time series section of hourly average mass concentration of BC, PM_{10} , long range transported dust factor and sum of traffic emissions and stationary combustion factors from 16 April 2010 to 23 April 2010 at the University of Applied Sciences in Augsburg, Germany.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



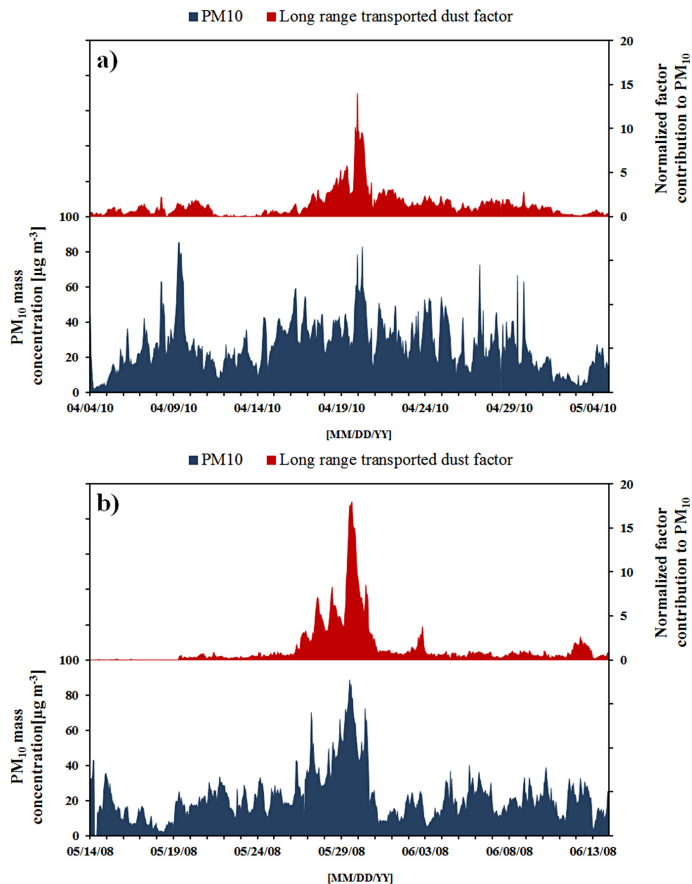


Fig. 5. Time series section of hourly average PM₁₀ mass concentration and normalized PM₁₀ contribution of long range transported dust factor during **(a)** Eyjafjallajökull ash plume impact 2010 and **(b)** Saharan dust event 2008 at the University of Applied Sciences in Augsburg, Germany.