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***Interactive comment on* “Ground-based and airborne in-situ measurements of the Eyjafjallajökull volcanic aerosol plume in Switzerland in spring 2010” by N. Bukowiecki et al.**

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We thank Reviewer 2 for her/his efforts and her/his fruitful comments, which we have fully considered in our revised version of the paper.

Our detailed answers are listed below:

General:

"It would be helpful to include an abbreviations list which would make it easier for the reader to follow the manuscript. There are a lot of abbreviations and not always are they fully written out the first time they are mentioned. A check of abbreviations is

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needed where they are only written out once and after this an abbreviations list will help the reader."

We have included a list of abbreviations as Appendix B

"A clarification of the terminology is needed to distinguish between volcanic ash and volcanic aerosols (volcanic ash + sulphate). This is not used consistently in the current manuscript version."

We have checked the manuscript for consistency regarding these expressions and have added the following statement to Section 3.1.1 (Identification of the volcanic plume): "Figure 2 shows that the volume size distributions measured during all these time periods with volcanic influence, exhibited a clear bimodality. The volcanic aerosol plume was thus characterized by an accumulation mode in the diameter range 0.1–0.8 μm and an ash mode (volcanic ash) with concentrations peaking around 3 μm . This terminology will be used throughout the rest of this paper."

Introduction:

"P12952, L 13-14: VAAC is not located in London. Please change to e.g. "...predictions by the London Volcanic Ash Advisory Centre (VAAC), which is part..."

ok

"P12953, L8: Please change "volcano plume" to "volcanic plume"."

ok

Methods:

"P12954, L24-25: Why is there a considerable loss of particles with $D > 15 \mu\text{m}$ in the sampling line?"

We have clarified this as follows: "Based on the length and geometric design of the inlet line, it is estimated that there is a considerable loss of particles with $D > 15 \mu\text{m}$ in

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the sampling line.”

"P12954, L26: Please clarify what polystyrene latex spheres (PSL) means and that volcanic ash is a non-PSL aerosol. It is stated that non-PSL aerosols will result in a diameter shift due to different refractive indices. It would be helpful to shortly explain what refractive index means physically and how and why this is different for PSL aerosols and volcanic ash particles. Also later in the manuscript the refractive index term is used substantially, so a short explanation of the physical meaning of the term will help readers not so familiar with particle physics to more easily understand this."

We have complemented the respective part in Section 2.2 as follows: “As described in detail in Appendix A, aerosol measurements with an OPC depend on the wavelength-dependent complex refractive index of the sampled aerosol, which determines the scattering response for a particle of a given size. The real part of the refractive index is mainly influenced by the scattering aerosol components, while the imaginary part varies with different contributions from absorbing species. As a result, the measurement of non-PSL aerosols like volcanic ash with a PSL-calibrated OPC will result in a diameter shift of the size distribution due to the different material specific refractive indices. This diameter shift has to be corrected for, because it has a strong influence on the calculation of volume and mass concentrations from the raw number size distributions.”

"P12955, L22: Section title “Analysis of air and snow samples by SEM, ICP-MS and IC” Please avoid abbreviations in section titles before they are explained."

We have changed the title to “Chemical and electron-microscopical analysis of air and snow samples”

"P12957, L2: Please change “volcano eruption” to “initial volcanic eruption” or “during the first phase of the volcanic eruption”."

ok

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"P12957, L9: Is it necessary to include the abbreviation for visual flight rules, VFR, which is not used further in the manuscript?"

We omitted the abbreviation

"P12958, L6: Did you use ECMWF forecast fields for the simulations? The simulation was performed after the eruption as an analysis exercise and only the reanalysis data would be used."

As mentioned in section 2.6 we used a mix of operational analysis (00, 06, 12, 18 UTC) and 3 hour forecast fields (03, 09, 15, 21 UTC) to achieve a 3-hourly resolution of meteorological input data. ERA Interim re-analysis fields are not used because they only provide forecasts at 00 and 12 UTC, not allowing constructing the desired temporal resolution of 3 hourly input data with minimal inconsistencies between the fields.

"P12958, L9-10: For the simulation only an ash mode particle of 3 μm was used. Maybe it is helpful to mention that this is the peak in the measured volume size distributions at JFJ. This will further strengthen the choice of only using this mode for the simulations. "

We added this information in section 2.6.

"P12958, L10: Is this 4% of the mass estimated by Stohl et al 2011, or of the total erupted mass from the volcano? The mass estimated by Stohl et al is not the total mass emitted from the volcano, but fine ash in the size range 2.8-28 μm . How did you determine that 4% of the mass was in the 3 μm mode?"

As mentioned also in the reply to referee 1, the 4 % given in the manuscript were a typo. It should read 40 %. These are 40 % of the mass emitted in the 2.8 – 28 μm size range as estimated by Stohl et al. 2011. The original assumption in Stohl et al. was that 10 % of the total emitted mass is in the size range mentioned. However, this was arbitrarily chosen and there is no evidence that this fraction is correct, since the inversion by Stohl et al. only determines the mass in the size range mentioned and not the total mass.

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So we clarified in the text that we used 40 % of the mass as determined by Stohl et al.. Considering the size distribution given in Stohl et al. this fraction seems to be too large, However, we tuned this fraction to best match the total mass observations in the coarse mode at Jungfraujoch, Zugspitze, and the Swiss NABEL sites.

Results and discussions:

"P12959, L20-26: It is not very obvious that the first peak in SO₂ and PM₁₀ when there was a southerly wind direction and low RH (late on 17th April) is connected to the volcanic ash. There is a subsequent drop in SO₂ which is mentioned but not elaborated further. This needs more discussion. How confident are you that this is volcanic ash even when SO₂ values drop? Did the FLEXPART simulation show any ash cloud from south coming in over the station?"

We have rephrased the respective paragraph and clarified the above issues: "The long-term measurements performed at the Jungfraujoch since 1995 suggest that normally no significant coarse mode aerosols are present with the exception of Saharan dust. . . . In April and May 2010 several episodes with coarse mode volume concentrations clearly exceeding the background levels were identified at the Jungfraujoch (Fig. 1a) beside the largely continuous presence of the accumulation mode The two events related to the volcanic plume were first recognized and characterized by strongly increased concentrations of PM₁₀ and SO₂ (Fig. 1b). As it will be discussed in Sect. 3.2.3, the Jungfraujoch only faced the diluted and rather inhomogeneous edge of the volcanic plume in April 2010. During the April event the volcanic plume reached the site for the first time under relatively dry conditions and local winds from South,, indicated by a simultaneous increase in PM₁₀ and SO₂. Subsequently, the local wind direction changed to NW, accompanied by an engulfment of the site in clouds The change in weather conditions coincided with a drop in SO₂, while PM₁₀ stayed constant. The changing levels of SO₂ are linked to changing oxidation and neutralization states of the volcano related sulfur, which is discussed in detail in Sect 3.1.2."

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We also added the following to Section 3.2.3. :”The FLEXPART simulation did not indicate any advection from the south. However, such local flow systems are unlikely to be represented in the employed meteorological fields and the simulations cannot be expected to reproduce such fine scale structures.”

”P12959, L26: Please discuss why there are two or three PM10 and SO2 peaks for the April event (one on 17th, 18th and 19th) while wind directions were rather constant. Could this reflect different emission pulses from the volcano?”

This is very difficult to state, we rather suspect the plume inhomogeneity to be the dominating reason (see reply to previous comment). The wind direction has to be considered to be very local, as later discussed in Section 3.2.3.

We have added the following statement to Section 3.2.3: ”FLEXPART did not simulate the arrival of considerable amounts of volcanic ash at the Jungfraujoch on 16 May. However, moderate amounts of ash were modeled to arrive from the North West on 17 May and the main peak observed on 18 May was only simulated for 19 May. The comparison suggests that there was a delay of about 6 to 12 hours in the simulations for the peak concentrations. Assuming that a similar delay was present in the simulations for 16 May, the conclusion that increased PM10 levels were due to volcanic ash is also backed up by the simulations”.

”P12959, L27: It is said that the plume was first detected at JFJ on 16 May. It is not obvious that these two small peaks in PM10 and SO2 are related to volcanic ash. The volume size distribution (Fig 1a) shows increased accumulation mode particles, which can suggest sulphate particles, but there is not a significant increase in the coarse mode particles for these two small peaks. Did your FLEXPART simulation show any ash over the station on the 16 May that can support this?”

While the mentioned increase is not very strong, it is still clearly above the normal coarse mode levels observed at the Jungfraujoch. To underline this we have added a respective statement to Section 3.1.1 (first phrase in reply to previous comment)

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"In the whole of this section it would be useful to try to distinguish between the increase in accumulation mode (suggesting sulphate particles?) and increase in coarse mode particles (suggesting volcanic ash?), and also to be clear in the terminology what you mean by volcanic aerosol plume/ volcanic influence. Are you talking about both sulphate and volcanic ash, or only the ash plume (on Figure 1 the events are marked as volcanic ash)? Please clarify."

We have rephrased the respective paragraph and clarified the above issues

"P12960, L3-5: Again it is not obvious that this small PM10 peak late on the 19th May is related to volcanic ash. Is it possible to see from the FLEXPART simulation whether an ash plume was transported in from south over the station at this particular time? To me it seems that the two main peaks in April and May have a significant increase in both the accumulation mode and the coarse mode particles. The small peaks in April and May do not clearly show this, with only a significant increase in either the accumulation or the coarse mode. As mentioned, a discussion on sulphate versus volcanic ash aerosols and clearly define volcanic aerosol plume may help this interpretation."

See, above: While the mentioned increase is not drastic, it is still clearly above the normal coarse mode levels observed at the Jungfraujoch. To underline this we have added a respective statement to Section 3.1.1. The FLEXPART simulations, even if somewhat delayed compared to the observations, support the conclusion that during the northfoehn event on 19 May large concentrations of volcanic ash are brought down to the surface in the lee of the Alps (Northern Italy) were subsequently the whole boundary layer fills up with volcanic ash. The secondary peak in the late afternoon of 19 May might then be a result of local flow systems bringing boundary layer air from the Valais or even the Po Valley back to the Jungfraujoch. Again such a flow would not be well captured by FLEXPART. However, it is interesting to note that the FLEXPART simulated age of the volcanic ash increased during the afternoon peak, suggesting that these air masses had another transport history than those responsible for the main peak.

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"P12959, L27: It is said that the plume was first detected again later on 19 May. I believe it is later on 18 May according to figure 1."

The reviewer is right, we have fixed this

"P12964, L7-8: "The imaginary part decreases indicating less absorbing (more transparent) particles", this is in contrary to P12963 L25-26 where you indicate presence of a significant portion of absorbing species within the volcanic ash coarse mode. Please clarify."

We have clarified this as follows: "The shown refractive indices have to be interpreted as a mean value for the entire size distribution and are strongly dominated by the accumulation mode. This dominance becomes apparent in the imaginary part, where the rather stable value of about 0.02i (average over all wavelengths) measured during the periods dominated by volcanic aerosol is much higher than the values estimated for the ash mode only. The clear decrease of the imaginary part during the volcanic aerosol plume indicates the higher relative contribution of transparent accumulation mode particles. In contrast the real part shows no significant change and illustrates that the scattering characteristics of the volcanic accumulation mode showed similar scattering characteristics like the normal background aerosol".

"P12964, L8: The real part shows no significant change (figure 10). This is not so obvious from the figure. It seems to increase for the maximum volcanic aerosol influence? What does the real part of the refractive index tell us physically and what does it mean if it has no significant change or if it increases?"

See our answer to the previous comment.

"P12965, L 6-7: The observed number concentrations on 18 April was lower than on the 17th for the aircraft measurements, which is opposite of the Jungfrauoch measurements. What can the reasons be? Didn't the aircraft "hit" the ash plume, or only on the very edge of it? Figure 14 and 15 show that on 17 April the aircraft was more in the

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centre of the plume."

FLEXPART simulated greater concentrations above the Swiss Plateau as compared to JFJ. The plume approaching from north-west was somewhat deflected by the Alps and therefore its greatest concentrations did not reach JFJ, which is situated within the northernmost main crest of the Swiss Alps. We have clarified this as follows:" While the stationary measurements at the Jungfraujoch did not capture the full volcanic plume (Sect 3.2.3), the aircraft measurements were useful to investigate the spatial plume inhomogeneity. ... The $N > 0.5$ number concentrations of the ash mode observed on the subsequent day (18 April 2010) were clearly lower compared to the values of the day before, indicating that the air masses containing the volcanic aerosol have further subsided."

"P12965, L14-16: The measured volume concentration on 18 May from DIMO was 5-10 times higher than the corresponding Jungfraujoch measurement (fig 12). What can be likely reasons for this? Was the aircraft closer to the centre of the volcanic plume (where is Biel and Emmental and how far is that from JFJ, from Figure 11 this is not seen)? Can you refer to the FLEXPART simulations to explain the difference in volume concentration from DIMO and JFJ to the locations of the measurements and whether they were taken more in the centre of the ash plume or at the edge (Figure 16 shows this)?"

See reply to previous comment

"P12966, L10-11: Do you mean the contribution of volcanic ash or volcanic aerosol (ash + sulphate)? Please define TiO_2 and why this can be used as a tracer for volcanic aerosol."

We have checked the paragraph for consistent terminology and added the following: "To estimate the mass contribution of volcanic ash to total PM_{10} at the involved stations, titanium dioxide (TiO_2) was used as suitable source specific tracer for the volcanic aerosol, because TiO_2 in PM_{10} was highly enriched during the volcanic ash

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episodes. TiO₂ in PM₁₀ in Switzerland is predominantly of geogenic origin and concentrations are typically low.”

"P12966, L19: “A value also supported by FLEXPART”. Is this shown on any figure? "

Yes, in Figure 15 (we have added the cross-reference)

"P12967, L7-10: Even though the weather conditions at Jungfraujoch was fairly stable, there are local meteorological conditions in the mountain area of the Swiss alps which will lead to a heterogeneous plume, however these local effects are often not very well captured by the meteorological input data to the model, and thus increased uncertainty of the modelled plume is to be expected."

We thank the reviewer for the suggestion, which we used as input.

"P12697, L13 and L24: Please change “the data” to “the model data”."

ok

Appendix A1:

"P12970, L14-15: It would be helpful to shortly describe what the real and imaginary parts of the refractive index tell us about the particles. Also, why are you varying the index parts in this range (real 1.4-16., imaginary 0i-0.005i)?"

A general description has been added to Section 2.2. Furthermore, we added the following sentence “These ranges include the values suggested in literature. . . and thus are expected to include also the effective values for the volcanic ash found at the Jungfraujoch.

"P12970, L15, and P12971 L9: Can you say “the Mie wiggles”, or better “the scattering cross section wiggles”?"

The expression “Mie wiggles” is used in regular literature (see e.g. Raymond, L.L., 1998, Mie theory, Airy theory, and the natural rainbow, APPLIED OPTICS 37(9), 1506-

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1519.

"P12971, L19: "as shown in Figure 12", and discussed in which section?"

Section 3.2.1 (we added the cross-reference)

Appendix A2:

"P12971, L27: Is it necessary to refer to Fig. 21 at this stage without more explanation?"

We think it's appropriate, because it shows the impact of the uncertainty estimate. The discussion takes place in the respective Section (3.2.1).

"P12972, L28: Please revise the unclear sentence: ", because the large and correction factor..""

We have rephrased the sentence

Figures:

"Figure 3: On the b panel the left axis should be coloured according to the line (black)."

We have fixed this

"Figure 4: Please specify all the chemical species shown in the figure."

We have specified the species but decided not to name out every individual chemical element in the caption

"Figure 11: It is not possible to see on a print version the labels of the marked cities. Can you also mark the location of Jungfrauoch? Either this figure needs to be revised with a larger right side panel, or the whole figure needs to be enlarged for the final version."

We have improved the readability and will ask the production office to insert this figure as a full page figure with portrait orientation. This will help to further improve the

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readability. The Jungfraujoch is not located in the shown map domain (this is now indicated in the caption).

"Figure 17: It is not clear from the figure what the effect of increasing imaginary part is as the line colors are the same for constant real part and changing imaginary part. Consider using the same line color and structure as in Figure 18. In the figure caption insert "Calculated Mie scattering..""

We have carefully checked the figure and improved the readability to an optimal extent

"Figure 18: Change the caption to "The OPC diameter correction" instead of "The Figures shows OPC.." Also it is said that "..for a complex refractive index of $1.54 + 0.005i$ ", but the imaginary part varies."

We have rephrased the caption to correct this.

"Figure 20: It is not possible to distinguish the blue lines with different symbols. Consider revising the figure with different line settings. Alternatively it could be stated that the differences are small and what the range is."

We have carefully checked the figure and improved the readability to an optimal extent

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12949, 2011.

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