Reply to reviewers comments on "Volcanic ash detection with infrared limb sounding: MIPAS observations and radiative transfer simulations"

General remarks

We thank the reviewers for their detailed comments. Both reviewers have provided thoughtful and helpful reviews that were taken into account in the revised version of the manuscript. In summary, we feel that we have addressed all the comments of the reviewers and that this has led to a much improved manuscript. We hereby confirm that all authors listed on the manuscript concur with submission of the paper in its revised form.

Below you find the original comments of the reviewers in *italics* and our response. Also the changes in the revised version of the manuscript in response of the comments received are shown in sans serif.

Reply to Reviewer 1

General Comments
I thought that this was a good paper which develops the volcanic ash "reverse absorption" technique to be applicable to limb sounders such as MIPAS. The authors use the Puyehue-Cardon Caulle eruption in 2011 to evaluate a detection threshold. A series of theoretical sensitivity experiments are conducted with ice, ash and sulphate aerosol refractive index data to evaluated the detection threshold. This provides a useful technique to detect and gain height information that will be useful when new limb instruments are launched to complement the information from nadir sounders.

I would recommend publication following the minor revisions highlighted in the sections below.

Specific comments
(1) pg 9941, line 2: the interest for climate work is emphasised but no mention is made for the impact on aviation. As this method provides information on the ash altitude it would provide useful additional information for volcanic ash advisory centres. A sentence should be added with a suitable reference

to detail the impact of ash on aviation. This point could also be added when discussing the ash altitude evaluations.

We added the following sentence to the introduction: Furthermore, volcanic ash poses a severe danger to aircraft (e.g. Casadevall, 1994). Hence, for aviation safety near real-time observations of volcanic ash plumes are essential.

And we added the following sentence to the discussion: Altitude-resolved information on volcanic plumes derived by utilizing this fast detection method could also be of use to e.g. the Volcanic Ash Advisory Centres.

(2) pg 9942, line 9: The authors say that this is "for infrared limb measurements so far no method has been reported that specifically allows for volcanic ash detection", however there is a recent paper which detects volcanic ash using MIPAS data (Grainger et al, 2013). I recommend removing the above sentence and other comments related to this.

We deleted this sentence and replaced it by: Concerning volcanic emission detection with MIPAS, two studies were recently published, Griessbach (2012a) discussed a method that specifically allows for volcanic ash detection and Grainger (2013) presented new methods for volcanic plume detection.

(3) pg 9945, line 18: the authors use the Volz 1973 refractive index data for volcanic dust. Why has this dataset been used? Why have other refractive index data not been considered? Is the Volz 1973 composition similar and comparable with the Puyehue-Cardon Caulle ash emitted in 2011? Volcanic ash composition is different for each volcano. As direct refractive index measurements for the Puyehue were not available, we decided to perform our study with the well-established Volz (1973) data set. This approach is also in agreement with Wen and Rose (1994), who found a stronger sensitivity on the size distribution than the refractive index for volcanic ash. However, we see the point in discussing uncertainties due to the refractive index and included our sensitivity tests now. We added further refractive indices to Figures 1-3 and also discussed it in Section 3.1.

Figure 1: We added the refractive indices of volcanic ash compounds measured by Pollack (1973) and modified the caption.

Section 3.1: We added a short discussion of these refractive indices and the corresponding extinction coefficient spectra. Paragraph 2: The real and imaginary parts of the complex refractive indices of ice (Warren and Brandt,2008), volcanic ash (Volz, 1973), volcanic ash components (andesite, basalt, basaltic glass, and obsidian Pollack et al., 1973), and sulphate aerosol (Hummel et al.,

1988) are shown in Figure 1 (a) and (b) for MIPAS band A. and All volcanic ash components have a larger refractive index at 950 cm^{-1} than at 825 cm^{-1}, but the amplitude of this difference varies.

Paragraph 3: The magnitude of the increase depends on the respective refractive index.

Paragraph 4: Regarding the single scattering albedo, the spectral slope shows a decrease similar to small ice particles for three volcanic ash refractive indices and for the other three volcanic ash refractive indices it is very similar to large ice particles (tropical cirrus).

Figure 2: We added the uncertainty ranges due to the refractive index for each particle size. The caption reads now: The extinction coefficient spectra are normalised at 826 cm^{-1}. The coloured areas denote the variation range due to different refractive index data sets for each size distribution. The black lines denote the Volz (1973) refractive index. The black area and the grey dashed line are calculated for the volcanic ash measurements reported by Schumann (2011b) with $r_{sca} = 3.7\,\mu$m.

Paragraph 5: The size dependency of ash particles and the variation range due to the refractive index is shown in Figure 2. and With increasing particle size the increase of the extinction coefficient from 825 to 950 cm^{-1} is getting smaller and disappears for a scattering radius of 4.9 μm. Also the variation range due to the refractive index is getting smaller with increasing particle size. For the size distributions with a scattering radius larger than 2.9 μm the differences due to the particle size are larger than the differences due to the refractive index. Only for the two smallest scattering radii the refractive index variation range is significantly larger than the differences due to particle size. However, the size distributions with the smallest scattering radii show the strongest spectral slopes, but also have the smallest single scattering albedos. Hence, from very small ash particle sizes we expect only a weak impact on the MIPAS spectra and ash particle size distributions with scattering radii larger than or equal to 4.9 μm will not be distinguishable from ice clouds for MIPAS band A spectra.

Figure 3: We added simulations for all volcanic ash refractive indices.

Paragraph 7: For both windows, at 825 and 950 $^{-1}$, we simulated the radiances for all volcanic ash types and compare them with a clear air and an ice cloud simulation in Figure 3a.

Paragraph 8: The differences in the extinction coefficient spectra for volcanic ash and the ice cloud directly transferred into the simulated spectra. When focusing on the spectral regions with the smallest trace gas contributions that are highlighted in grey, it is clearly visible that all volcanic ash types lead to a significant increase in radiance at 950 $^{-1}$ and the ice cloud leads to a decrease in radiance at 950 $^{-1}$ compared to clear air. For this particular size distribution the Volz (1973) refractive indices caused the smallest radiance increase compared to the other refractive indices and hence is the most conservative assumption.

(4) pg 9946, line 16: the particle size distribution has a constant width of 1.6 why was this value chosen? How sensitive is the detection to significant changes in this value?

Volcanic ash size distributions can have widths ranging from 1.1 to 2.2 according to Püschel et al. (1994). Widths measured by Farlow (1981) range from 1.5 to 1.7. We chose 1.6, because it is in the middle. To explain the reason for the chosen width, we added the following sentence:Because volcanic ash size distributions have widths ranging from 1.1 to 2.2 (Farlow et al. 1981, Pueschel et al.,1994) we chose a constant width of 1.6, which is in the middle.

For our simulations we always calculated r_{eff} and r_{sca}. Both depend on the median radius and the width of the size distribution. When keeping the median radius constant and changing the width, r_{eff} and r_{sca} also change. Hence, the effect of a varying width is covered by the variations of the scattering radius. For particle sizes between $0.1-10\,\mu$m around $10\,\mu$m wavelength the scattering radius is the best measure to account for dependencies on the particle size distribution. As shown in Section 3.1 Figure 2, the measured (3-modal) volcanic ash size distribution is best represented by the (monomodal) model size distribution with an equivalent scattering radius and not by the size distribution with an equivalent effective radius. Hence, for a better reproducibility we also added the scattering radius to Tables 3, 4, and 5.

(5) pg 9948, line 10: 12.2 km seems a very specific height and I couldn't find this information from the web link given. Perhaps "around 12 km" is more appropriate.

On the web page in the Section 1 June-7 June 2011 we find the following sentences: "Later, an explosion from Cordón Caulle produced a 5-km-wide ash-and-gas plume that rose to an altitude of 12.2 km (40,000 ft) a.s.l. as noted by OVDAS scientists." "Based on analyses of satellite imagery, SIGMET notices, and information from the Puerto Montt Flight Information Region (FIR), the Buenos Aires VAAC reported that on 4 June ash plumes rose to altitudes of 10.7-13.7 km (35,000-45,000 ft) a.s.l. and drifted 870 km ESE." "The next day an ash plume continued to rise to altitudes of 10.7-12.2 km (35,000-40,000 ft) a.s.l. and had drifted as far as 1,778 km ESE, over the coast of Argentina, and out into the Atlantic Ocean." Therefore we rephrased our text: about 11 − 14 km altitude

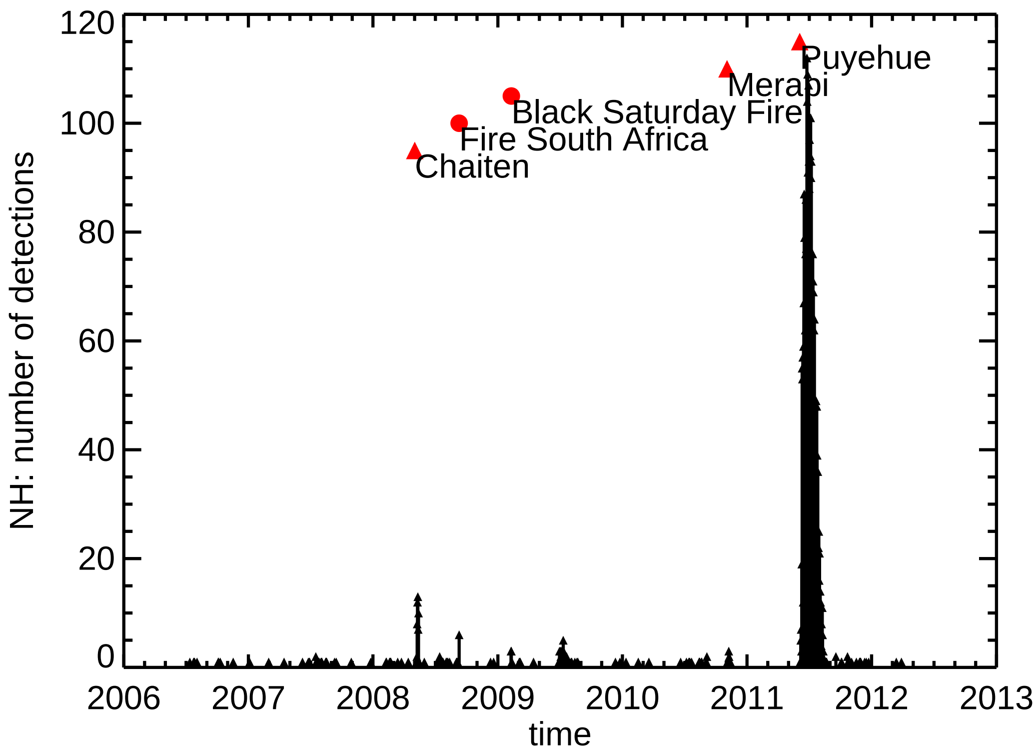
(6) pg 9950, line 19: the authors state that we expect to find volcanic ash in this region and give references. If the MIPAS detection was compared (in figure 7) to a SEVIRI disc (ash product or ash/dust rgb available from eumetsat) or a composition of MODIS or IASI images this would provide more compelling evidence to support the conclusion that MIPAS has successfully detected volcanic ash.

We replaced Figure 7 by 2 new figures that show MIPAS detections and an AIRS ash index for a 12 h time window. The new caption reads now: MIPAS and AIRS volcanic ash detections between 12 and 24 UTC on 7 and 9 June 2011. The AIRS ash index (brightness temperature(960.7 cm^{-1}) - brightness temperature(833.2 cm^{-1})) is colour coded. The black circles indicate the profiles where volcanic ash was detected by MIPAS. The Puyehue-Cordón Caulle is marked by the red triangle and the black dashes denote the MIPAS tangent points below 30 km.

We also modified the last paragraph of Section 3.3: In Figure 7, the Puyehue-Cordón Caulle volcanic ash plume detected by MIPAS and AIRS between 12 and 24 UTC on 7 and 9 June 2011 are shown. On 7 June the AIRS data show the ash plume confined to a rather narrow filament extending from south Brazil over the south Atlantic to south of the southern tip of Africa. Two days later the ash plume was transported further east and a wave-like filament extends from the south Atlantic to the south Pacific. Some fresher plumes are located over the Atlantic close to the South American coast. MIPAS detects the volcanic ash in the same regions as AIRS. Slight differences in location can be attributed to differences in measurement time (up to 12 hours), as the plume is moving very fast. Furthermore, we also processed the MIPAS data from 2006 to 2012 and detected volcanic ash after all major eruptions, such as Kasatochi, Chaiten, and Sarychev (Griessbach,2012a).

(7) Could the authors include some statistics on the number of false alarms and failed detection of volcanic ash.

The Figure below shows the number of profiles with a volcanic ash detection per day from 2006 to 2013 in the southern hemisphere. Per day about 650 profiles are measured in the southern hemisphere. We indicated the major events that we have reliably identified so far. Unfortunately we cannot give precise numbers, because for the whole MIPAS measurement period (2002-2012) global measurements with a similar high sensitivity to aerosol in the UTLS are not available. To do comparisons with other instruments, e.g. IASI, AIRS, CALIOP, forward and backward trajectory calculations would be required for each detection to allow for comparisons at the same time. This is far beyond the scope of our study. Regarding failed detections, MIPAS does not measure below approx. 5 km in the polar region and 10 km in the tropics. Below these altitudes ash plumes may be captured by nadir instruments, but not by MIPAS.



(8) Could the authors include some details on the geolocation of the Tangent point. As accurately locating where the ash is located is vital for aviation.

Mainly the information arises from a 30×300×3 km (across track×along track×vertical) box around the tangent point. We added this information to Section 2.2:The field of view at the tangent point extends about 3 km in the vertical direction and 30 km perpendicular to the line of sight. The dimension of the measurement volume along the line of sight is about 300 km.

(9) Fig 1, Fig 6, Fig 8, Fig 9, fig 10, Fig 13 are difficult to interpret because of the grayscale used. These would be much clearer if they were in colour.

Done.

(10) Fig 2: Why is 3.7 at the top of the legend rather than in the order set below it? Please change or add an explanation to the figure 2 caption.

3.7 is the scattering radius for the only measured size distribution used in this figure. The figure caption was revised, please see answer to comment (3).

Technical corrections

(1) pg 9940, line 2: change "troposphere" to "the troposphere" and "stratosphere" to "the stratosphere".

Done.

(2) pg 9941, line 8: reword sentence beginning "Especially" to something like: "Infrared emission measurements are especially useful as they provide day and night observations".

Done.

(3) pg 9941, line 11: add Prata 1989a as well to the Prata reference.

Done.

(4) pg 9941, line 13: add e.g. before Barton et al. as the list of papers provided is notan exhaustive list.

Done.

(5) pg 9941, line 19 to 25: this sentence is too long and hard to follow, consider re-wording and breaking in several smaller sentences.

It reads now: Analyses of space-borne and balloon borne infrared limb aerosol measurements focus on the detection of stratospheric volcanic sulfate aerosol, but do not aim at volcanic ash detection or cloud and aerosol classification in the upper troposphere. Stratospheric volcanic sulfate aerosol has been observed from space by the cryogenic limb array etalon spectrometer (CLAES) (Massie,1996b; Lambert,1997 and the improved stratospheric and mesospheric sounder (ISAMS) (Grainger,1993; Lambert,1993), both aboard the Upper Atmospheric Research Satellite (UARS). Also the balloon-borne Michelson Interferometer for Passive Atmospheric Sounding (MIPAS-B) measured stratospheric volcanic sulfate aerosol during a campaign in Kiruna in 1992 (Echle,1998).

(6) pg 9942, line 1: replace "to detect" with "the detection of"

Done

(7) pg 9942, line 1: remove "Consecutively" because its unnecessary

Done

(8) pg 9942, line 12: change "in particular MIPAS" to "using MIPAS data" and remove the comma.

Done

(9) pg 9944, line 4: change "index i" to "index i, respectively"

Done

(10) pg 9945, line 3: change "works" to "work"

Done

(11) pg 9946, line 24: change "equal 5" to "equal to 5"

Done

(12) pg 9950, line 13: "This does not change the result" at all or significantly?

Changed to: "This did not change the results significantly."

(13) pg 9951, line 26: remove first and and replace with comma

Done

(14) pg 9952, line 10: change first sentence to "Wen and Rose (1994) found a stronger sensitivity on the size distribution than the refractive index for volcanic ash therefore the microphysical properties are restricted to one refractive index dataset per particle type and the particle size distribution is varied."

Done

(15) pg 9954, line 7: change "equal 6" to "equal to 6"

Done

(16) pg 9958, line 11: change strongest for largest

Done

(17) pg 9958, line 11: change "yr of Mt." to "years, Mt."

Done

(18) pg 9958, line 17: change oxidated to oxidised.

Done

(19) pg 9958, line 18: change over to in

Done

(20) pg 9958, line 26: remove last sentence and add "giving confidence in our ash detection methodology" to the end of the previous line.

Done

(21) pg 9958, line 28: remove the before sulphuric acid.

Done

(22) pg 9958, line 29: replace having with have.

Done

(23) pg 9959, line 16: change from to using.

Done

(24) pg 9959, line 25: add shown after eruption.

Done

(25) pg 9960, line 10: change "also is" to "is also"

Done

(26) pg 9960, line 11: smoke? Can you be more specific, black carbon forest fires?

We changed it to: "... or smoke from wildfires."

(27) pg 9962, line 10: change "to estimate the layer bottom altitude" to "the layer bottom altitude to be estimated"

Done

(28) pg 9962, line 24: change sentence beginning "These windows" to something like "These window channels can be used to discriminate ash and meteorological cloud because of the spectral gradient produced by different particulates."

We rephrased it to: "These windows can be used to discriminate volcanic ash and ice clouds, because of characteristic spectral gradients produced by different particles."

(29) pg 9963, line 21 sentence beginning "We found" needs re-wording.

Done. It reads now: "Our simulations showed that background aerosol as well as volcanically enhanced sulfate aerosol, as measured after e.g. the Pinatubo eruption, can explicitly be discriminated from volcanic ash."

(30) Fig 7: consider making the black dots smaller for clarity. Change text from "with volcanic ash detections" to "where volcanic ash was detected"

Done

(31) Fig 8: define th in caption or change "tangent altitudes" to "tangent heights (th)"

Done

References: Grainger, R. G., et al. "Measuring Volcanic Plume and Ash Properties from Space." Remote-sensing of Volcanoes and Volcanic Processes: Integrating Observation and Modelling, edited by: Pyle, DM, Mather, TA, and Biggs, J., The Geological Society Special Publication 380 (2013).

Reply to Reviewer 2

In their paper Griessbach et al. present a technique for the fast detection of volcanic ash clouds from mid-infrared limb emission spectra. Based on extensive radiative transfer simulations and examples from MIPAS/Envisat observations it is shown that volcanic ash can be detected and distinguished from ice and sulphate aerosols within certain limits of size and extinction coefficients. The paper provides a valuable baseline for further in-depth investigations of volcanic eruptions from limb-emission observations. Further it clearly demonstrates the complementarity of limb-sounding with its high sensitivity and altitude resolution and nadir measurements with their large horizontal resolution. The manuscript is well written and clearly structured and I strongly recommend publication within AMT after a few clarifications/corrections as mentioned below.

Specific comments
p. 9940, l. 17 'derived the detectable effective radius range of 0.2 to 3.5μm': I strongly doubt that there exists a lower size limit: in the mid-IR the scattering from particles smaller than about 0.2-0.5 um is generally small compared to the absorption. Thus, the resulting IR spectra are no more dependent on the aerosol size, but only on the aerosol volume density (volume absorption; i.e. only on the imaginary part of the refractive index). This means that a spectrum from 0.2 m large particles looks the same as one with 0.01 m particles, under the condition that the total aerosol volume is constant. Since the spectral feature of volcanic ash, which is the baseline for the detection method, is mainly due to the imaginary part (as shown in Fig. 1) and as

mentioned in the text, it is most pronounced for small particles. Thus, I cannot see the need for a lower size limit.

From the theoretical point of view it is true that 0.2 μm is not the lower detection limit. However, to reach detectable extinction coefficients (volumes) with particles of this size an extremely high particle concentration is required. To our knowledge, such a high particle concentration has not been reported in measurements. To be clear, we changed this sentence in the abstract to: 'From the simulations we derived the upper detectable effective radius of 3.5 μm and the detectable extinction coefficient range of 5e-3 to 1e-1 km^{-1}.

In Section 4.4 we changed in paragraph 2: For particle size distributions with effective radii of 0.2 μm and smaller very high particle concentrations (more than 2000 cm^{-3}) are required to reach detectable extinctions. Although there is no physical reason for a lower detection limit, we think that it is unlikely to detect volcanic ash particle size distributions with effective radii smaller than 0.2 μm with this method, because such high particle concentrations have not been reported in measurements.

And we also clarify this point in the conclusions in more detail: We further found that for volcanic ash particles with effective radii smaller than 0.2 μm the particle concentrations must be unrealistically high to be detectable. Hence, we conclude that these small ash particles are very unlikely to be detected with this method.

p. 9940, l. 25 'Because they are efficient scatterers of ultraviolet and infrared radiation (Pueschel et al., 1994), they change the aerosol optical thickness': In the infrared, they are at least similar strong absorbers of IR radiation. This should be made clear.

We added: ".. (Pueschel,1994) as well as good absorbers in the infrared, they change.."

p9945, l. 18 'volcanic ash (Volz, 1973)': Also other sources of volcanic ash refractive indices should be given. Further, I strongly recommend that these are shown, e.g. in Fig. 1, and some simulations be performed, e.g. for the example of Fig. 3. Otherwise the whole study is based on one example of refractive indices which makes the case a bit weak.

We added other volcanic ash compound refractive indices and simulations. For details please see answer to comment 3 of Reviewer 1.

p9945, l.28 'The spectral slope of the imaginary part of the refractive indices directly determines the extinction coefficient spectra': Doesn't it depend also *strongly on the particle size via the real part?*

Yes, we deleted this sentence to avoid a longer general description and now focus even more on discussing the simulation results. For details please see answer to comment 3 of Reviewer 1.

p.9946, l.11 'Therefore we expect a weaker radiance increase for sulphate aerosol in the MIPAS spectra than for ice and volcanic ash.': A weaker increase with respect to what? To aerosol mass?

We added the missing information: "Therefore we expect a weaker radiance increase relative to clear air for sulfate aerosol than for ice and volcanic ash in the MIPAS spectra."

p.9947 and Fig. 3:

1. The exact position of the tangent points should be stated.

Done.

2. In Fig. 3b, for the ice cloud simulation a subvisible cirrus has been used with rather small particles. However, I think profile 18 of MIPAS orbit 48509 is located at northern mid-latitudes which makes the use of SVC not obvious. (Particularly, further below on p. 9954 it is mentioned that: 'These median radii are very small and can only be expected in the tropics for sub-visible cirrus clouds.')

Subvisible cirrus occurs quite frequently at northern mid-latitudes at altitudes between 10-11 km. A statistics from SAGE II measurements can for example be found on Plate 2 in Gierens et al. (2000). We chose a subvisible cirrus as an example, because it provides the largest contrast. In Section 3.1, paragraph 7, we rephrased the following sentence: For volcanic ash, the particle size distribution with the steepest extinction coefficient slope between both windows for the Volz (1973) refractive indices, i.e. effective radius of 1 μm, was taken and for the ice cloud, the most contrasting particle size distribution of the SVC was taken. And on p. 9954 we deleted "in the tropics", because it is not correct there.

Gierens et al.: Ice-supersaturated regions and subvisible cirrus in the northern and southern mid-latitude upper troposphere, JGR, Vol 105, No D18, 22743-22753, 2000

p.9948, l.3: It is mentioned that in the simulation for Fig. 3a, the water vapour has been assumed too high compared to the measurements in Fig. 3b. Can you indicate how much of the radiance continuum is due to the water *vapour continuum.*

We added the following sentences: The differences between volcanic ash simulations with and without water vapour are 2 and 5 percentage points at 825.5 and 950.5 cm^{-1} respectively. As the impact of water vapour on the ash cloud spectra is very small and nearly constant, it does not affect the spectral slope due to volcanic ash.

Technical comments

p.9952, l.27 and p.9995, l.3: check units (W (cm2...))

Done. The radiance units are always $\mathrm{W(m^2\ sr\ cm^{-1})^{-1}}$. There is no p.9995.

Fig.1: Lines are very difficult to distinguish. Use color?

Done.

Figs 8-10, 13: Use color to distinguish plot symbols.

Done.

F. 12, caption: Units for the extinction coefficients are missing.

We added the unit.