Responses to Reviewers - amt-2018-447

I would like to begin by thanking the reviewers and editor for their time taken to read and comment on the manuscript titled 'An adaptation of the CO_2 slicing technique for the Infrared Atmospheric Sounding Interferometer to obtain the height of tropospheric volcanic ash clouds' (amt-2018-447). We believe that these comments have helped to improve the content and clarity of the manuscript and we hope that you agree.

To summarise, the main changes to the text include:

- **Section 4.1.** Further detail on why the ranges of AOD and ER were used within this study for channel selection.
- **Section 4.1.** We've expanded the description of figure 3 to make this easier for the reader to understand.
- **Section 5.2.** Further explanation of the differences between the CO_2 slicing and optimal estimation scheme results. This includes an expanded description of the OE scheme (specifically focused around the a priori) and why this version of the OE does not perform as well as the CO_2 slicing in these cases.

Summary of changes to figures:

- **All →** We have changed the font type and increased the font size of all the plots to make them easier to interpret.
- **Figure 2.** Y axis labelled corrected to hPa. Figure caption has been updated to indicate the channels used in this example.
- **Figure 3 (and equivalent appendix plots).** Channel combinations not explored in this analysis has been shaded in an off white to distinguish these from channel combinations which failed for all cases. X axis label has been added.
- **Figure 4.** Channel combinations not explored in this analysis has been shaded in an off white to distinguish these from channel combinations which failed for all cases. X axis label has been added.
- **Figure 6 (and equivalent appendix plots).** Three additional panels have been included which show the frequency of AOD/effective radius/pressure for which the CO_2 slicing returned a value which was within 0.5 km of the simulation height. These help support conclusions that the technique performs less well for smaller AOD/ER and at the pressure extremes tested.
- **Figure 8.** Legend added for the scatterplots.
- **Figure 9.** The error bars have been replaced with shaded polygons. The shaded region indicates the standard deviation away from the mean height which has been explained in the figure caption.
- **Figure 10-13.** Panel e has been removed.

Additional changes have been made to improve clarity of the writing and to avoid misunderstandings. Where identified changes have been made to spelling and typing errors. Individual comments are addressed below. The reviewer's comments are coloured in blue and are in bold font. Our responses are offset from these and in black. Text in italics are relevant passages from the revised text. Where possible we indicate the line numbers of the altered text. All changes can be seen in the track changes of the manuscript which can be found at the end of this document.

Response to Anonymous Referee #1

The words "meteorological cloud" is used a lot. It is not obvious what that term means. Some explanation should be given to introduce it.

To avoid confusion, we now use the words "aqueous clouds" and define this when it is mentioned in the introduction **(now page 2, lines 22-23)**:

'Similar retrieval techniques exist to obtain the cloud height of aqueous clouds (i.e. water/ice clouds not associated with volcanic activity)'

Line 12, page 3: "Assuming an atmosphere which is decreasing in temperature with height": This is not a realistic assumption since it is true only for the troposphere. There is some discussion on this in the last paragraph of section 2 but it should probably be moved here.

This has been rephrased **(now page 3, lines 11-12)**:

'In the Earth's troposphere where temperature is decreasing with height, the radiances measured by the instrument are proportional to the transparency of the atmosphere for each channel'.

Do the simulated spectra cover the range of atmospheric conditions expected over the globe? The authors use six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter. What about tropical night, summer and winter in the tropics and mid-latitudes, etc?

We believe that the six atmospheres chosen captured sufficient atmospheric variability to demonstrate the applicability of the method. From our results the method is weakest as the atmospheric lapse rate approach zero (polar summer). As the suggested atmospheres are far from isothermal, we do not believe much would be learned given the computational expense.

Lines 6-7, page 6: Need a reference(s) showing that those are appropriate values for ash cloud properties.

In this study we have used a range simulated ash spectra. For the channel selection we use ash spectra representative of optically thick volcanic ash clouds (AODS: 5-15, ER: 5-10). We then test this on a wider range of ash properties which represent thinner plumes before applying the technique to real ash scenes. We have expanded the first paragraph in section 4.1 to indicate why we chose these ranges and to emphasise that it is then tested on a wider range of properties including those more representative of thinner ash clouds:

'IASI has over 300 channels which fall within the CO_2 absorption band, and so, to ensure computational efficiency an appropriate subset of these channels must be selected. To do this the CO_2 slicing technique was first applied to 384 simulated ash spectra. These are `ideal' test cases which do not include other aerosols or aqueous cloud. These spectra include six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter (including atmospheric profiles created for MIPAS; Remedios et al., 2007). The spectra were modelled using the refractive indices of samples of volcanic ash from the Eyjafjallajökull eruption in 2010 (Peter, 2010): the main eruption considered in this study. In the future different refractive indices could be used such as those in Prata et al. (2019). A range of ash properties were explored: cloud heights between 200 and 900 hPa (going slightly above the tropopause), ash effective radius between 5 and 10 μm, and ash optical depths between 5 and 15

(referenced at 550 nm). Typically, the effective radius is less than 8 µm for very fine ash (such as in a distal plume) and between 8 and 64 µm for fine ash (Marzano et al. 2018). The range of ash optical depths is highly variable. Ventress et al. (2016) and Balis et al. (2016) recorded ash optical depths of less than 1.2 from dispersed plumes from Eyjafjallajökull in 2010; however much higher values can be expected closer to the volcano or following large explosive eruptions. The effective radius and AOD explored here for the channel selection is in the upper range and above what might be expected: values which may only be true close to the volcanic vent. The spectrum of an optically thin plume is more difficult to differentiate from a clear spectrum commonly leading to the signal ($I_{obs}(v)$-$I_{clr}(v)$) to be within the instrument noise and subsequently will result in no retrieval. A decision was made to select the channels used using idealised optically thick cases, which may only be true close to the vent, for which the plume should be evident in the majority of the CO_2 channels. The selected channels are tested on a wider range of AODs and effective radius in section 4.2 including smaller values that are more representative of a disperse plume.' **(page 6, lines 3-21)**

How is the weighting function w = d(tau)/d(lnp) computed?

We compute the derivative of tau (output from RTTOV) with logarithm of pressure for the CO_2 channel using the IDL deriv routine. This gives weighting function profiles such as those seen in figure 1d.

The use of the weighting function (now k) to obtain a single pressure solution was originally mentioned on page 7, line 16. We've expanded this sentence to explain what this means and where the value for τ comes from **(now page 8, lines 4-5)**:

'... where p_c is the pressure retrieved for channels v and k refers to the weighting function based on the derivative of atmospheric transmittance computed for each pressure level with RTTOV with respect to the log of atmospheric pressure ($d\tau[v,p]/d\ln p$).'

Fig. and Figure are both used. Choose one and be consistent.

We have followed the convention outlined by AMT in the Manuscript preparation guidelines. We have used Fig. where it appears in the running text and Figure at the start of a sentence.

It is surprising that a technique using just a few channels (CO_2 slicing) outperforms one that uses many more channels and retrieves several parameters self-consistently using radiative transfer simulations and iterative fits to spectra (OE). The authors suggest that this may be due to the OE retrievals being strongly influenced by the height a priori. This may be so. However, this suggests that the measurements do not have much information on ash height (otherwise, the prior should not strongly affect the retrievals). If that is the case, how does the CO_2 slicing obtain a better retrieval? A qualitative discussion of the difference between the two techniques is in order (not just that the results are different, but why they are different).

In this study, we have compared the CO_2 slicing results against the height output from an optimal estimation scheme, the results of which have been published previously (Ventress *et al.* 2016). This optimal estimation technique uses 105 channels, 14 of which are within the CO_2 absorption band. The channels used were not selected for their ability to obtain the ash cloud height and the previous study acknowledged that this is something that could be improved. Where there is not sufficient information about the height within the channels then the output would tend to the prior. Changes could be made to the OE retrieval, such as the inclusion of further channels within the CO_2 absorption band and this might improve the results. In this case however, we are comparing our results against the previously published study.

To avoid misleading the reader, we have removed the statement saying that the '*CO_2 slicing technique performs better than the OE technique*' (previously **page 1, line 13**) as re-reading this, this might imply that the CO_2 slicing method performs better than *any* optimal estimation scheme rather than just the version chosen for comparison.

We have also reworded the discussion of why the output of the two retrievals is different and improved the description of the a priori:

'By contrast, the OE average heights are less variable: between 3 and 4.25 km throughout the period studied. Some example maps of the OE results are shown in Fig. 10 to 13. The different assumptions and limitations of the two techniques mean that it is not expected that the two retrievals will return the same or even similar values. The optimal estimation scheme uses only 105 channels between 680.75 and 1204.5 cm^{-1} (~8.3 - 14.6 µm) to improve computational efficiency. This includes 14 channels within the CO_2 *absorption band, only one of which is in common with the* CO_2 *slicing. However, unlike the* CO_2 *slicing method presented here, the channels used by the optimal estimation scheme have not been optimised for retrieving the height of the ash layer. Ventress et al. (2016) noted that the optimal estimation retrieval could be further refined by altering the channels used. For example, channels with more height information could be selected. Similarly, Ventress et al. (2016) suggested that channels could be selected to minimise the effect of the underlying cloud layers following observations that the OE method can underestimate the cloud top height in cases of multiple cloud layers (Ventress et al. 2016). In the current application of the optimal estimation scheme, where there is not sufficient information about the height of the ash layer within the channels used, the retrieval height output will tend to the a priori height which in this case is around 3.5 km. This is potentially the reason for the persistently lower average height shown in Fig. 9 which suggests a strong dependence on the a priori.'* **(page 11, line 26 - page 12, line 7)**

<u>Technical Comments</u>

Line 7, page 2: which can result is -> which can result in

Now page 2, line 4. This has been corrected.

Line 10, page 3: need reference for RTTOV

This has been added:

'This has been simulated with the fast radiative transfer model RTTOV (version 9, Saunders et al. 1998) …' **(page 3, lines 7-8)**

Line28, page 4: remove "That"

Now page 4, line 27. Done

Line 30, page 4: "The second" -> "The third"

The original passage read: *'(2) the two channels used in Eq. 1 are sufficiently close that the difference in emissivity between them is negligible; (3) in cases where there are multiple layers of cloud, the lower level clouds are ignored. The second is particularly important to consider when the channel pairs are selected.'*

The channel pairs selected must have a negligible emissivity difference – this is referring to the second stated assumption. This paragraph has been restructured to avoid confusion:

'The CO_2 slicing method makes a number of assumptions: (1) the cloud is infinitesimally thin; (2) in cases where there are multiple layers of cloud, the lower level clouds are ignored; (3) the two channels used in Eq. 1 are sufficiently close that the difference in emissivity between them is negligible: this is particularly important to consider when the channel pairs are selected. Multiple cloud layers have previously been identified as a source of error in the CO_2 slicing retrieval …' **(page 4, lines 27-30)**

Line 8, page 5: dependant -> dependent

Now page 5, line 7. Done

Line 1, page 9: demonstrated -> demonstrates

Now page 10, line 4. Done

Line 23, page 9: including the CO_2 slicing technique -> including those obtained using the CO_2 slicing technique

This has been reworded:

'CALIOP and other LiDAR instruments are commonly used as a tool for the validation of cloud heights, including previous studies with the CO_2 *slicing technique'* **(page 10, line 24-26)**

Figure 3 caption: lines of the plot -> rows of the plot

Changed.

Figure 6 caption: The plots show the true (simulated) pressure plotted against the CO_2 slicing retrieved value for the six different atmospheres. -> Panels (a)-(f) show the true (simulated) pressure plotted against the CO_2 slicing retrieved value for the six different atmospheres.

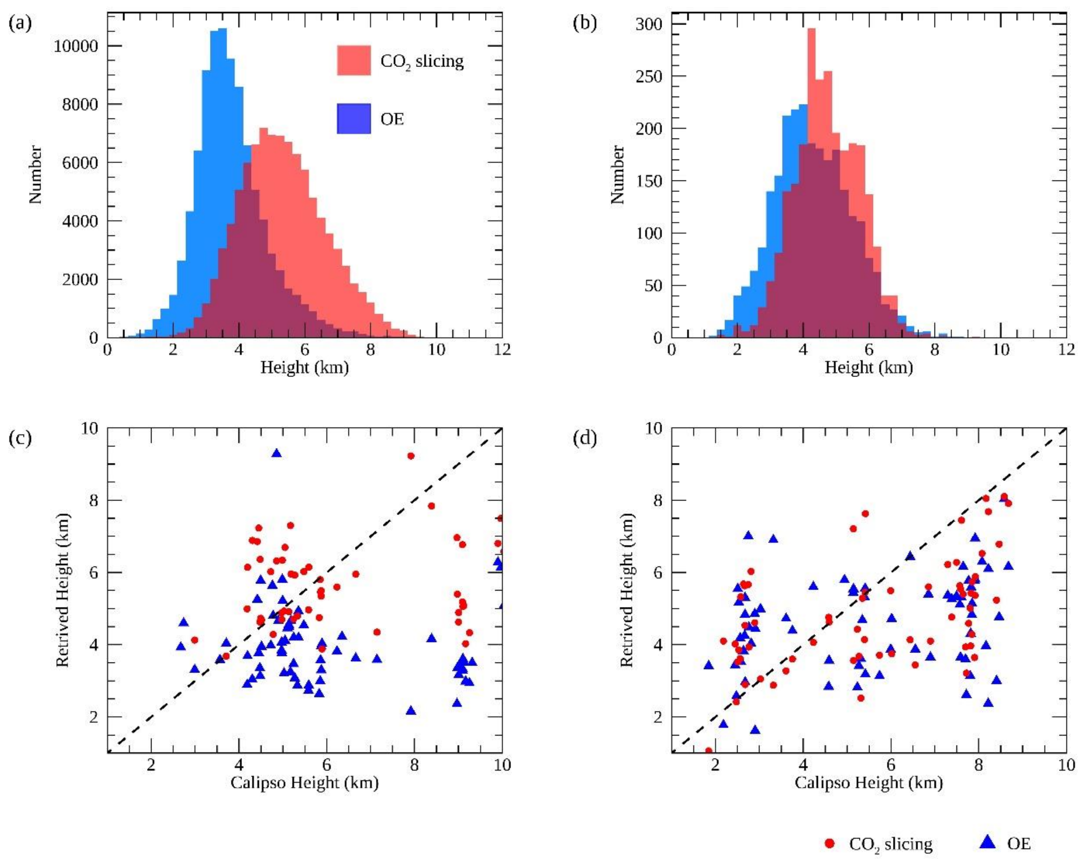
Changed

Figure 8 caption: The authors should note that the maroon distribution represents CALIOP retrievals

Panels (a) and (b) of figure 8 show the distribution of the CO_2 slicing (red/maroon) and optimal estimation (blue) heights obtained for the Eyjafjallajökull and Grímsvötn eruptions respectively. This is for all the pixels to which the retrieval was applied. Neither shows the distribution of heights obtained from CALIOP.

Panels (c) and (d) show scatterplots comparing the heights obtained with CALIOP (x axis) with those retrieved by the two IASI retrievals (CO2 slicing in red and OE in blue) again for both eruptions.

We've expanded the figure caption to make this easier to understand and added a legend for panels (c) and (d):



'(a) Distribution of the CO_2 slicing and optimal estimation retrieved ash heights for all pixels from the Eyjafjallajökull eruption. (b) Same as (a) for the Grímsvötn eruption. (c) Comparison of the CALIOP heights with those obtained with the CO_2 slicing and optimal estimation techniques for a subset of pixels (where measurements fell within 50 km and 2 hours of each other) from the Eyjafjallajökull eruption. (d) Same as (c) for the Grímsvötn eruption. Related statistics can be seen in table 5.'

Response to Anonymous Referee #2

Major Comments

1) Abstract and elsewhere: The statement in the abstract reads "Overall, the CO_2 slicing tool performs better than the optimal estimation scheme", which stopped me in my tracks! In common with the report of Referee #1, I was surprised by this – why indeed should the CO_2 slicing approach (which is basically a cut-down version of a full OE retrieval) outperform the OE approach? I feel that this should be explored in some more detail, because this (for me) is the main scientific issue in this manuscript. The authors suggest that the prior height constraint is responsible for the low ash height bias shown by the OE retrievals – to me, this suggests that the prior is clearly not appropriate in this case, or is being given far too much weight in the analysis.

Following here the answer to the same question posed by referee #1:

In this study, we have compared the CO_2 slicing results against the height output from an optimal estimation scheme, the results of which have been published previously (Ventress *et al.* 2016). This optimal estimation technique uses 105 channels, 14 of which are within the CO_2 absorption band. The channels used were not selected for their ability to obtain the ash cloud height and the previous study acknowledged that this is something that could be improved. Where there is not sufficient information about the height within the channels then the output would tend to the prior. Changes could be made to the OE retrieval, such as the inclusion of further channels within the CO_2 absorption band and this might improve the results. In this case however, we are comparing our results against the previously published study.

To avoid misleading the reader, we have removed the statement saying that the '*CO_2 slicing technique performs better than the OE technique*' (previously **page 1, line 13**) as re-reading this, this might imply that the CO_2 slicing method performs better than *any* optimal estimation scheme rather than just the version chosen for comparison.

We have also reworded the discussion of why the output of the two retrievals is different and improved the description of the a priori:

'By contrast, the OE average heights are less variable: between 3 and 4.25 km throughout the period studied. Some example maps of the OE results are shown in Fig. 10 to 13. The different assumptions and limitations of the two techniques mean that it is not expected that the two retrievals will return the same or even similar values. The optimal estimation scheme uses only 105 channels between 680.75 and 1204.5 cm^{-1} (~8.3 - 14.6 μm) to improve computational efficiency. This includes 14 channels within the CO_2 *absorption band, only one of which is in common with the* CO_2 *slicing. However, unlike the* CO_2 *slicing method presented here, the channels used by the optimal estimation scheme have not been optimised for retrieving the height of the ash layer. Ventress et al. (2016) noted that the optimal estimation retrieval could be further refined by altering the channels used. For example, channels with more height information could be selected. Similarly, Ventress et al. (2016) suggested that channels could be selected to minimise the effect of the underlying cloud layers following observations that the OE method can underestimate the cloud top height in cases of multiple cloud layers (Ventress et al. 2016). In the current application of the optimal estimation scheme, where there is not sufficient information about the height of the ash layer within the channels used, the retrieval height output will tend to the a priori height which in this case is around 3.5 km. This is potentially the reason for the persistently lower average height shown in Fig. 9 which suggests a strong dependence on the a priori.'* **(page 11, line 26 - page 12, line 7)**

2) Section 4.1, paragraph beginning "Figure 3 demonstrates. . .": I felt that this paragraph didn't really do justice to the description of Figure 3, and I think it would be good if the reader could be "guided" through the details of this Figure a little bit more. When you say ". . .the best performing channels. . .", do you mean CO_2 channels, reference channels, or both? When you say "As expected, this shifts from lower wavenumbers at lower pressures to higher wavenumbers closer to the surface", I found it very hard to interpret the intended meaning. Is "this" referring to the "best performing channels"? I really couldn't reconcile this sentence with what I was seeing in Figure 3.

The description of figure 3 was previously on **page 6, lines 24-34**.

We have expanded this paragraph to provide more detail on what is shown in figure 3. We now hopefully guide the reader through this better and demonstrate that the height of the ash layer affects the performance of the different channel combinations:

'Figure 3 demonstrates that the best performing channel pairs vary depending on the height of the plume. For plumes at lower pressures, the maximum pressure difference between the simulated and retrieved pressures is smaller at lower CO_2 *wavenumbers. For example, for the plumes simulated at 300 hPa, the maximum pressure difference was lowest (less than 20 hPa) for* CO_2 *channels between 700 and 710 cm*$^{-1}$*. As the pressure of the ash layer is increased, values are no longer obtained at smaller wavenumbers. For example, for a plume at 500 hPa, solutions are no longer obtained for* CO_2 *channels which are less than 700 cm*$^{-1}$*: the maximum pressure difference between the true and retrieved values is now smaller for slightly higher wavenumbers. For a plume at 800 hPa the maximum pressure difference is lowest (less than 60 hPa) for* CO_2 *channels between 715 and 720 cm*$^{-1}$*. This observation reflects what is shown in figure 1b and c: that the channel's peak sensitivity shifts from higher in the atmosphere at lower wavenumbers to close to the surface as higher wavenumbers effecting the best performing channel combination. Notably, at 200 hPa there...'* **(page 7, lines 5-14)**

3) Section 5.2, page 10, 28-29: You suggest that "In future applications of the OE scheme, the CO_2 slicing results could be used as the a priori". I disagree very strongly with this statement! One CANNOT use as prior information a state which has already been influenced by the measurements themselves. You could use the CO_2-slicing solution as the first guess in the OE iterative process, but absolutely not as the prior constraint. There's an equivalent comment in Section 6 (line 16 on page 12).

The optimal estimation scheme used for comparison in this study uses a total of 105 channels as opposed to the entire spectra to improve computational efficiency. Of these only 14 are within the CO_2 band. The optimal estimation scheme only uses one channel which is also used by the CO_2 slicing scheme. If that channel were excluded from the optimal estimation retrieval, then the CO_2 slicing result could be used as an a priori as it is not based on the same measurements. This has hopefully been clarified with the addition of further description about the optimal estimation scheme (as described in response to your first comment) and an additional line has been included after the comment about using the CO_2 slicing heights as an a priori in section 6:

'In future applications of the OE scheme, the CO_2 *slicing results could be used as the a priori if the one* CO_2 *channel that the two retrievals have in common was removed from the optimal estimation scheme'* **(page 12, lines 7-9)**

Minor comments, grammar, typos, suggestions, etc.

1) Section 2, page 4, line 2: "dependant" in this context should, I think, be "dependent"?

Now page 3, line 30. Done

2) Section 3, page 5, line 16: Trivial I know, but EUMETSAT usually insist that the satellite name is "Metop" and not "MetOp"!

Now page 5, line 15. Done

3) Section 4.1, and elsewhere: I note that you use "mb" for pressure units – I suspect the journal would prefer "hPa".

This has been changed throughout.

4) Section 4.1, page 6, line 9: When you use the phrase "is greater than the CO_2 channel", in what sense is "greater than" meant in this context? Channel number, wavenumber? Best to be absolutely explicit for clarity.

This was referring to wavenumber which has now been stated:

'The CO_2 slicing method was first applied using every channel combination between 660 and 800 cm^{-1}, where the reference channel (v_2) wavenumber is greater than the CO_2 channel (v_1) wavenumber.' **(page 6, lines 22-23)**

5) Section 4.2, page 6, line 16: When you say "The top two lines show. . .", I suggest using the word "rows" instead of "lines".

Now page 6, line 31. Done

6) Section 4.2, page 6, lines 26 and 29: You use the phrase "less channels" a couple of times. It should be "fewer channels".

Now page 7, lines 14 and 17. Done

7) Section 4.2, page 7, lines 16-19: It's not made clear in the text why you make the distinction between using narrower ranges for the channel selection work (ash optical depths ranging between 5 and 15, ash effective radius between 5 and 10 microns) than for the simulated retrieval work (ash optical depths ranging between 0.5 and 15, ash effective radius between 1 and 10 microns). It would be good for you to be explicit as to exactly why you didn't use "spectra representative of thinner ash clouds" for the channel selection.

Following here the answer to the same question posed by referee #1:

In this study we have used a range simulated ash spectra. For the channel selection we use ash spectra representative of optically thick volcanic ash clouds (AODS: 5-15, ER: 5-10). We then test this on a wider range of ash properties which represent thinner plumes before applying the technique to real ash scenes. We have expanded the first paragraph in section 4.1 to indicate why we chose these ranges and to emphasise that it is then tested on a wider range of properties including those more representative of thinner ash clouds:

'IASI has over 300 channels which fall within the CO_2 absorption band, and so, to ensure computational efficiency an appropriate subset of these channels must be selected. To do this the CO_2 slicing technique was first applied to 384 simulated ash spectra. These are `ideal' test cases which do not include other aerosols or aqueous cloud. These spectra include six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter (including atmospheric profiles created for MIPAS; Remedios et al., 2007). The spectra were modelled using the refractive indices of samples of volcanic ash from the Eyjafjallajökull eruption in 2010 (Peter, 2010): the main eruption considered in this study. In the future different refractive indices could be used such as those in Prata et al. (2019). A range of ash properties were explored: cloud heights between 200 and 900 hPa (going slightly above the tropopause), ash effective radius between 5 and 10 µm, and ash optical depths between 5 and 15 (referenced at 550 nm). Typically, the effective radius is less than 8 µm for very fine ash (such as in a distal plume) and between 8 and 64 µm for fine ash (Marzano et al. 2018). The range of ash optical depths is highly variable. Ventress et al. (2016) and Balis et al. (2016) recorded ash optical depths of less than 1.2 from dispersed plumes from Eyjafjallajökull in 2010; however much higher values can be expected closer to the volcano or following large explosive eruptions. The effective radius and AOD explored here for the channel selection is in the upper range and above what might be expected: values which may only be true close to the volcanic vent. The spectrum of an optically thin plume is more difficult to differentiate from a clear spectrum commonly leading to the signal ($I_{obs}(v)$-$I_{clr}(v)$) to be within the instrument noise and subsequently will result in no retrieval. A decision was made to select the channels used using idealised optically thick cases, which may only be true close to the vent, for which the plume should be evident in the majority of the CO_2 channels. The selected channels are tested on a wider range of AODs and effective radius in section 4.2 including smaller values that are more representative of a disperse plume.' **(page 6, lines 3-21)**

8) Section 5, page 8, line 31: "planck" should have upper-case "P".

Now page 9, line 30. Done

9) Section 5, page 9, line 33: You say that you have defined the tropopause "as the height at which the temperature profile inverts and has a positive gradient", but it's confusing that the tropopause dashed lines in Figures 6(a-f) have obviously not used this definition!

This has been rewritten to avoid confusion. When applying the CO_2 slicing technique to simulated data to select the channels and then test the applicability of the technique, the procedure was allowed to return values above the tropopause to test how successfully it performed. As the method was shown to perform poorly when the temperature gradient is stable, for the application to real ash scenes, it was only allowed to retrieve up to the tropopause as defined by the WMO. Figure 6 shows the tropopause (dashed line) as defined by the WMO.

The rewritten passage:

'Another point to note is that, in section 4, the maximum height that could be retrieved was defined as the height at which the temperature profile inverts and has a positive gradient. This is slightly above the tropopause which is defined by the World Meteorological Organisation (WMO) as the point at which the lapse rate is less than 2°C/km, and remains lower than this for at least 2 km. This was done to demonstrate how the CO_2 slicing method performs above the troposphere where the atmospheric temperature does not vary significantly: the atmospheric lapse rate here approaches zero. Figure 6 demonstrates that the CO_2 *slicing method performs poorly in these cases and so in the application to real data the* CO_2 *slicing method is only allowed to retrieve values up to the tropopause as defined by the WMO.'* **(page 9, line 32 - page 10, line 6)**

10) Section 5, page 9, lines 1-2: When you say "Figure 6 demonstrated that the CO_2 slicing method performs poorly where the temperature profile steepens significantly", can you clarify exactly what "steepens" means in this context – when dealing with (negative) vertical gradients, it's very easy for the reader to become confused with words such as "steepens"!

This has been rephrased to improve clarity:

'This was done to demonstrate how the CO_2 slicing method performs above the troposphere where the atmospheric temperature does not vary significantly: the atmospheric lapse rate here approaches zero.' **(page 10, lines 2-4)**

11) Section 5.2, page 10, lines 31-32: You say that "Ventress et al. (2016) identified that in some cases the retrieval assumed a lower altitude and a higher ash optical depth in order to fit the spectra". Lower/higher than what? Just needs to be a little clearer.

We have expanded this sentence:

'Ventress et al. (2016) identified that in some cases the retrieval underestimated the altitude of the plume and obtained a high ash optical depth in order to fit the measured spectra, when in reality the ash layer might have a lower optical depth and higher altitude.' **(page 12, lines 11-13)**

12) Figure 4: What is the x-axis here? The y-axis is labelled simply as "Wavenumber". Presumably they should both be the same as for Figure 3?

The x axis was also wavenumber. The axes have now been labelled CO_2 wavenumber (x axis) and reference wavenumber (y axis).

13) Figures 10-13: Where does the retrieved ash mass come from – the OE retrievals? In any case, it's not clear to me exactly why these mass column loadings have been included in the paper, as I don't believe they are ever referred to. What do they add to the paper?

The ash mass can be calculated from the ash optical depth and effective radius obtained with the optimal estimation scheme (assuming an ash density). The maps of these are included for reference.

We have included an additional line within the manuscript which makes reference to these plots:

'Some example maps of the OE heights are shown in Fig. 10 to 13b, alongside the ash mass (panel c) calculated from the OE retrievals of AOD and effective radius, assuming an ash density. The maps of ash mass show that in general the ash mass falls with transportation away from the vent.' **(page 11, lines 27-30)**

As we do not refer to panel (e) of figures 10-13 within the paper we have decided to remove these.

14) Table 3: Needs units! Presumably the "Channel Ranges" etc. refer to wavenumbers (cmˆ-1)? Are the "Peak Sensitivity Ranges" in mb/hPa?

We've added units to the table. The channel ranges/reference channel are in wavenumber (cm^{-1}) and the peak sensitivity was in (hPa).

Response to Anonymous Referee #3

Specific Comments:

In the description of simulated ash spectra (section 4.1, page 6, lines 6-7), are specified the AOD, Effective Radius and Cloud Heights ranges used, but no reference about the ash type considered, that is the aerosol optical properties (extinction and absorption coefficient, asymmetry parameter, etc . . .) used in simulation. Which ash type was used? Andesite? Obsidian? Pumice? Other?

An additional line has been included:

'The refractive index used in this study is from measurements made of ash from the Eyjafjallajökull eruption (Peters, 2010): the main eruption considering in this study. In the future different refractive indices could be used such as those in Prata et al. (2019).' **(page 6, lines 7-9)**

Why you use different AOD and Effective Radius ranges for channel selection (section 4.1) and simulation results (section 4.2)? AOD=5-15 and Ref=5-10 micron for channel selection, AOD=0.5-15 and Ref=1-10 micron for simulation results. Can you explain better?

Following here the answer to the same question posed by referee #1:

In this study we have used a range simulated ash spectra. For the channel selection we use ash spectra representative of optically thick volcanic ash clouds (AODS: 5-15, ER: 5-10). We then test this on a wider range of ash properties which represent thinner plumes before applying the technique to real ash scenes. We have expanded the first paragraph in section 4.1 to indicate why we chose these ranges and to emphasise that it is then tested on a wider range of properties including those more representative of thinner ash clouds:

'IASI has over 300 channels which fall within the CO_2 absorption band, and so, to ensure computational efficiency an appropriate subset of these channels must be selected. To do this the CO_2 slicing technique was first applied to 384 simulated ash spectra. These are `ideal' test cases which do not include other aerosols or aqueous cloud. These spectra include six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter (including atmospheric profiles created for MIPAS; Remedios et al., 2007). The spectra were modelled using the refractive indices of samples of volcanic ash from the Eyjafjallajökull eruption in 2010 (Peter, 2010): the main eruption considered in this study. In the future different refractive indices could be used such as those in Prata et al. (2019). A range of ash properties were explored: cloud heights between 200 and 900 hPa (going slightly above the tropopause), ash effective radius between 5 and 10 µm, and ash optical depths between 5 and 15 (referenced at 550 nm). Typically, the effective radius is less than 8 µm for very fine ash (such as in a distal plume) and between 8 and 64 µm for fine ash (Marzano et al. 2018). The range of ash optical depths is highly variable. Ventress et al. (2016) and Balis et al. (2016) recorded ash optical depths of less than 1.2 from dispersed plumes from Eyjafjallajökull in 2010; however much higher values can be expected closer to the volcano or following large explosive eruptions. The effective radius and AOD explored here for the channel selection is in the upper range and above what might be expected: values which may only be true close to the volcanic vent. The spectrum of an optically thin plume is more difficult to differentiate from a clear spectrum commonly leading to the signal ($I_{obs}(v)$-$I_{clr}(v)$) to be within the instrument noise and subsequently will result in no retrieval. A decision was made to select the channels

used using idealised optically thick cases, which may only be true close to the vent, for which the plume should be evident in the majority of the CO_2 channels. The selected channels are tested on a wider range of AODs and effective radius in section 4.2 including smaller values that are more representative of a disperse plume.' **(page 6, lines 3-21)**

I think it would be very interesting to evaluate the heights obtained from CO_2 slicing as a function of AOD, Ref and cloud top pressures. In fig. 6 (g-i) are shown the frequency for which the CO_2 slicing was unable to return a height value. I suggest to insert 3 similar panel to show the frequency for which the CO_2 slicing returns a good value (for example a value that differ from the truth max +/- 500 meters or +/- 1 km). In this way we could better understand in which conditions the CO_2 slicing is applicable and reliable.

As suggested, three additional panels were added to figure 6 which is shown below. These show the frequency of cases where the difference between the simulated and retrieved values are less than 0.5 km. These plots demonstrate that the CO_2 slicing technique performs slightly less well for cases with smaller ash optical depths and effective radius. It also supports the previous observation that the CO_2 slicing technique does not perform as well at the pressure extremes tested.

We have included the updated plot in the manuscript and added a few lines in section 4.2:

'[about failed aods and er] …. This observation is supported by figure 6j-k which shows the number of cases where the difference between the simulated and retrieved pressure is less than 0.5 km: which is slightly lower for a smaller effective radius and ash optical depth.' **(page 8, lines 19-21)**

'[about failed extreme pressures] …. The majority of failed cases are shown to be at the pressure extremes, Fig. 6i. Similarly, Fig. 6l indicates that there are fewer cases where the pressure difference between the simulated and retrieved pressures are less than 0.5 km at these pressures.' **(page 8, lines 22-23)**

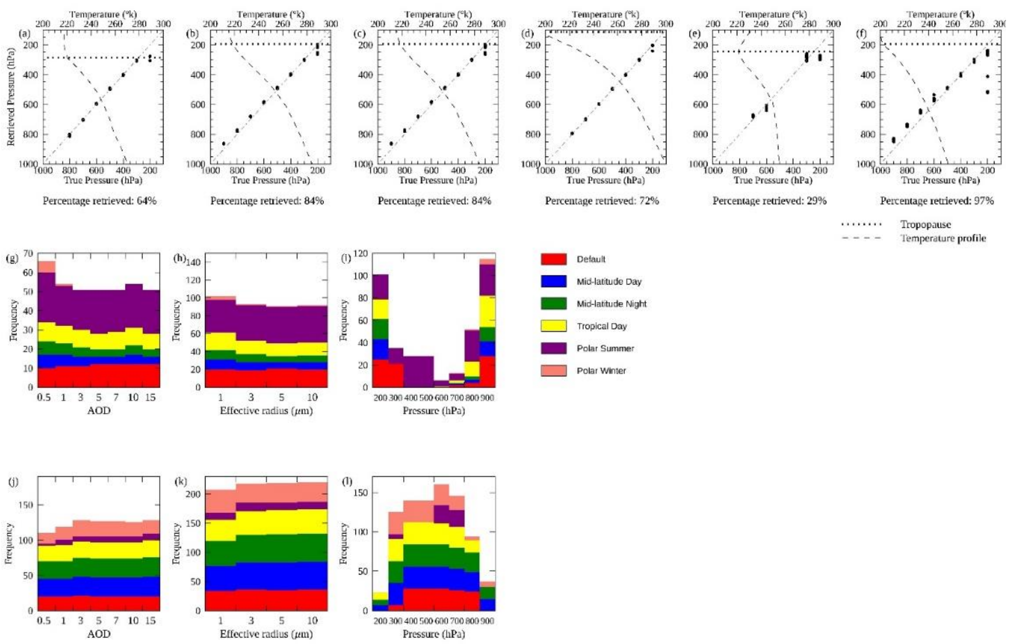


Figure 6 caption: Final CO_2 slicing pressure results for RTTOV simulated ash spectra (a total of 224 spectra per atmosphere). Panels (a)-(f) show the true (simulated) pressure plotted against the CO_2 slicing retrieved value for the six different atmospheres. (a) RTTOV default atmosphere (high latitude), (b) Mid-latitude day, (c) Mid-

latitude night, (d) Tropical day, (e) Polar summer (f) Polar winter. In this case, the simulated spectra include the following ash properties: ash optical depth ranging between 0.5 and 15, ash effective radius ranging between 1 and 10 µm and pressure values between 200 and 900 hPa. Below each plot is a value indicating the percentage of successful retrievals (where a height value can be obtained and all quality control conditions have been met). (g) The frequency of ash optical depths for which the CO_2 slicing technique was unable to return a height value. (h) Same as (g) for the effective radius. (i) Same as (g) for the ash cloud pressure. (j) The frequency of ash optical depths for which the difference between the simulation and CO_2 slicing height is less than 0.5 km. (k) The same as (j) for effective radius. (l) The same as (j) for ash cloud pressure. Related statistics can be seen in table 4. The equivalent plot, where the values which have not met the quality control conditions has been included in the appendix, figure A7.

Technical Comments

Page 4, line 1: transmittance is not radiance, so it can't be "emitted" . . .. I suggest to replace the sentence with: "the atmospheric transmittance at channel v of the layer between the pressure level p and the instrument (top of atmosphere)".

Now page 3, lines 29-30. Done

Page 5, line 3: "with" instead of "which" ?

We've changed which to that. The line now reads: *'The effect of surface emissivity is expected to be minimal as channels within the* CO_2 *absorption band have weighting functions that peak above the surface, as shown in Fig. 1d.'* **(page 5, lines 1-3)**

Page 10, line 6: here you said "4 days" for Grímsvötn eruptions, while in fig. 7 the days are only 3 (20110521 PM, 20110522 AM, 20110522 PM, 20110523 AM).

Now page 11, line 8. Corrected.

Figure 2: the y-label is "Pressure (mbar)" not "Altitude (km)".

Updated to hPa.

Figure 3: the x-label of the last two lines is missing ("CO_2 Wavenumber (cm-1)").

This has been corrected.

Figure 4: the same as above.

This has been corrected.

Responses to Referee #4

Specific Comments

Page 3 line 10: Add version of RTTOV.

This now reads:

'This has been simulated with the fast radiative transfer model RTTOV (version 9, Saunders et al. *1998) and replicates what would be observed with IASI given specified atmospheric conditions'* **(page 3, lines 7-8)**

Page 3 line 28: The expression "L_obs v_1" should be replaced to "L_obs (v_1)".

Now page 3, line 27. Done

Page 4 line 7: "w" is used for window channel in Eq. (3) but later it uses for weighting function.

The weighting function has been changed to k

Page 4 line 9: There are no explanation for L_cld (v) in the text. Is it the same as L_obs (v)?

Now page 4, line 8. Where it occurs $L_{cld}(v)$ has been replaced with $L_{obs}(v)$.

Section 4: In the approach of CO_2 slicing of this paper, contribution of meteorological clouds seems to be omitted in radiance calculations. If so, it should be mentioned in the text.

We've added an additional line to clarify:

'To do this, the CO_2 *slicing technique was first applied to 384 simulated ash spectra. These are 'ideal' test cases, which do not include other aerosols or aqueous cloud.'* **(page 6, lines 4-5)**

Page 6 line 5-6: Add reference for the atmospheric profiles.

A reference has been added for the MIPAS atmospheric profiles:

'These spectra include six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter (including atmospheric profiles created for MIPAS; Remedios et al. 2007)' **(page 6, lines 5-7).**

The reference for these profiles: https://www.atmos-chem-phys-discuss.net/7/9973/2007/

Page 6 line 6-7: Add the applied ash model of refractive index for ash optical properties. Is it Pollack andesite model?

An additional line has been included:

'The refractive index used in this study is from measurements made of ash from the Eyjafjallajökull eruption (Peters, 2010): the main eruption considered in this study. In the future different refractive indices could be used such as those in Prata et al. (2019).' **(page 6, lines 7-9)**

Page 6 line 13-14: The values or reference for the noise of the instrument should be added.

We've added this:

'(1) $L_{obs}v_1$-$L_{clr}v_1$ must be greater than the noise of the instrument at channel v_1 (CO_2 channel within the CO_2 *absorption band the noise of the IASI instruments is between 2.55×10^{-8} and 3.77×10^{-8} $W/(cm^2.sr.cm^{-1})$)'* **(page 6, lines 27-29)**

Page 8 line 28-29: More explanation for the flagged pixel is required. Do you determine the flagged pixels by yourself? What channel and threshold value are used? If the flagged pixels were given from somewhere, the data source should be added.

More detail has been added to describe the method used to flag the ash pixels prior to the application of the CO_2 slicing and optimal estimation techniques:

'In this application of the retrieval, it has only been applied to pixels which are flagged as containing volcanic ash by a linear ash retrieval developed for IASI (Ventress et al. 2016; Sears et al. 2013): following the method developed for SO_2 by Walker et al. 2012). This method compares each IASI spectra against a covariance matrix formed from pixels which contain no volcanic ash thereby representing the spectral variability associated with interfering gas species or clouds, and also the instrument noise. A least squares fit is performed for three ash altitudes (400, 600 and 800 hPa) to retrieve a value for ash optical depth. A pixel is then flagged if it exceeds a threshold at any height. As SO_2 can, with caution, be used as a proxy for volcanic ash (Carn et al., 2009; Thomas and Prata, 2011) the retrieval has also been run for pixels flagged for SO_2 using the same approach (Walker et al. 2011, 2012; Carboni et al. 2012, 2016).' **(page 9, lines 20-27)**

Page 8 line 29-30: Add description of surface condition (temperature and emissivity) for the calculations of L_clr. Did you use the surface emissivity model in RTTOV?

We've added a few lines to explain this:

'*For the* CO_2 *slicing values for L_{clr} were obtained using the radiative transfer model RTTOV using the ECMWF atmospheric profile as an input and using the default ocean emissivity within RTTOV. The effect of surface emissivity is thought to be minimal as for the channels used the weighting functions peak above the surface, Fig. 1d.'* **(page 9, lines 27-30)**

Page 9, Sec 5.1.1: Detailed explanation for the determination of the a priori ash height in the optimal estimation scheme is needed in the text. It is the important point in the discussion for the results of comparison between OE and your CO_2 slicing.

We have expanded the discussion of the OE scheme within section 5.2 where the results of the OE and CO_2 slicing techniques are compared. Within this we give more detail on why the CO_2 slicing performs better in these cases and why the OE is affected by the a priori used.

Following here the answer to a similar question posed by referee #1:

In this study, we have compared the CO_2 slicing results against the height output from an optimal estimation scheme, the results of which have been published previously (Ventress *et al.* 2016). This optimal estimation technique uses 105 channels, 14 of which are within the CO_2 absorption band. The channels used were not selected for their ability to obtain the ash cloud height and the previous study acknowledged that this is something that could be improved. Where there is not sufficient information about the height within the channels then the output would tend to the prior. Changes could be made to the OE retrieval, such as the inclusion of further channels within the CO_2 absorption band and this might improve the results. In this case however, we are comparing our results against the previously published study.

To avoid misleading the reader, we have removed the statement saying that '*the CO_2 slicing technique performs better than the OE technique*' (previously **page 1, line 13**) as re-reading this, this might imply that the CO_2 slicing method performs better than *any* optimal estimation scheme rather than just the version chosen for comparison.

We have also reworded the discussion of why the output of the two retrievals is different and improved the description of the a priori:

'By contrast, the OE average heights are less variable: between 3 and 4.25 km throughout the period studied. Some example maps of the OE results are shown in Fig. 10 to 13. The different assumptions and limitations of the two techniques mean that it is not expected that the two retrievals will return the same or even similar values. The optimal estimation scheme uses only 105 channels between 680.75 and 1204.5 cm^{-1} (~8.3 - 14.6 µm) to improve computational efficiency. This includes 14 channels within the CO_2 *absorption band, only one of which is in common with the* CO_2 *slicing. However, unlike the* CO_2 *slicing method presented here, the channels used by the optimal estimation scheme have not been optimised for retrieving the height of the ash layer. Ventress et al. (2016) noted that the optimal estimation retrieval could be further refined by altering the channels used. For example, channels with more height information could be selected. Similarly, Ventress et al. (2016) suggested that channels could be selected to minimise the effect of the underlying cloud layers following observations that the OE method can underestimate the cloud top height in cases of multiple cloud layers (Ventress et al. 2016). In the current application of the optimal estimation scheme, where there is not sufficient information about the height of the ash layer within the channels used, the retrieval height output will tend to the a priori height which in this case is around 3.5 km. This is potentially the reason for the persistently lower average height shown in Fig. 9 which suggests a strong dependence on the a priori.'* **(page 11, line 26 - page 12, line 7)**

Figure 2: Label of the ordinate seems wrong. Add values of v_1 and v_2 in this calculation.

Figure 2 has been updated to show pressure on the y axis. We've added the following line to the caption to indicate which channels are used:

'In this example v_1 and v_2 are at 715 cm^{-1} and 725 cm^{-1} respectively.'

Figure 10-13: There are no discussions for the plots of ash mass (e). Add discussion if these plots are important.

These panels in figures 10-13 have been removed.

Table 3: What does the number of "step" in table 3 mean? Why step 2 does not exist?

This column in table 3 has been removed.

Figure A7: In the caption of Fig. A7, same sentences as those of Fig.6 is not necessarys

The caption has been edited and now reads:

'Same as figure 6 without a quality control applied.'

An adaptation of the CO_2 slicing technique for the Infrared Atmospheric Sounding Interferometer to obtain the height of tropospheric volcanic ash clouds

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Abstract. Ash clouds are a geographically far reaching hazard associated with volcanic eruptions. To minimise the risk that these pose to aircraft and to limit disruption to the aviation industry, it is important to closely monitor the emission and atmospheric dispersion of these plumes. The altitude of the plume is an important consideration and is an essential input into many models of ash cloud propagation. CO_2 slicing is an established technique for obtaining the top height of meteorological aqueous clouds and previous studies have demonstrated that there is potential for this method to be used for volcanic ash. In this study, the CO_2 slicing technique has been adapted for volcanic ash and applied to spectra obtained from the Infrared Atmospheric Sounding Interferometer (IASI). Simulated ash spectra are first used to select the most appropriate channels and then demonstrate that the technique has merit for determining the altitude of the ash. These results indicate a strong match between the true heights and CO_2 slicing output with a root mean square error (RMSE) of less than 800 m. Following this, the technique was applied to spectra obtained with IASI during the Eyjafjallajökull and Grimsvötn eruptions in 2010 and 2011 respectively, both of which emitted ash clouds into the troposphere, and which have been extensively studied with satellite imagery. The CO_2 slicing results were compared against those from an optimal estimation scheme, also developed for IASI, and a satellite borne LiDAR is used for validation. Overall, the CO_2 slicing tool performs better than the optimal estimation scheme. The CO_2 slicing heights returned a RMSE value of 2.2 km when compared against the LiDAR. This is lower than the RMSE for the optimal estimation scheme (2.8 km). The CO_2 slicing technique is a relatively fast tool and the results suggest that this method could be used to get a first approximation of the ash cloud height, potentially for use for hazard mitigation, or as an input for other retrieval techniques or models of ash cloud propagation.

1 Introduction

Encounters of aircraft with volcanic ash have demonstrated that such occurrences can cause significant damage to the plane (Casadevall, 1994; Dunn and Wade, 1994; Pieri et al., 2002; Guffanti and Tupper, 2015). In extreme cases, these have resulted in engine failure (Miller and Casadevall, 2000; Chen and Zhao, 2015) and subsequently life-threatening circumstances. Ash clouds are closely monitored by the Volcanic Ash Advisory Centres (VAACs) who use a variety of data sources including information from volcano observatories and satellite data (Prata and Tupper, 2009; Thomas and Watson, 2010; Lechner et al., 2017). This allows informed decisions on the closure of airspace following an eruption, which can result ~~is~~ in severe disruption and have significant financial implications. For example, the eruption of Eyjafjallajökull in 2010, resulted in the closure of a large portion of Northern European airspace and subsequently, the cancellation of 100,000 flights and a revenue loss of $1.7 billion (IATA Economic Breifing, 2010). Alongside these potential impacts to the aviation industry, volcanic ash is also a hazard to health (Horwell and Baxter, 2006; Horwell, 2007) and can cause considerable damage to infrastructure (Durant et al., 2010; Wilson et al., 2012, 2015).

Satellite remote sensing, particularly infrared instruments, has been widely used for monitoring the hazards presented by volcanic ash. This has included detection schemes which flag pixels that contain volcanic ash (e.g. Prata, 1989a, b; Ellrod et al., 2003; Pergola et al., 2004; Filizzola et al., 2007; Clarisse et al., 2010; Mackie and Watson, 2014; Taylor et al., 2015). Other methods have been developed to quantify parameters such as the mass, ash optical depth (AOD), effective radius and altitude of the ash cloud, usually relying on look up tables or optimal estimation techniques (e.g. Wen and Rose, 1994; Yu et al., 2002; Watson et al., 2004; Corradini et al., 2008; Gangale et al., 2010; Francis et al., 2012; Grainger et al., 2013; Pavolonis et al., 2013).

Knowing the position of the ash cloud in three dimensions is critical for hazard mitigation. Plume height is a crucial part of this and it is also a variable in models of ash cloud propagation (Mastin et al., 2009; Stohl et al., 2011; Bonadonna et al., 2012) such as HYSPLIT (Draxier and Hess, 1998; Stein et al., 2015) or NAME (Jones, 2004; Witham et al., 2012). A number of different methods have been used to obtain the height of volcanic ash clouds. These have included the use of ground based and airborne instruments, and satellite techniques (Glaze et al., 1999), some of which are summarised in table 1.

This problem is not unique to volcanic ash. Similar retrieval techniques exist to obtain the cloud height of ~~meteorological clouds~~ aqueous clouds (i.e. water/ice clouds not associated with volcanic activity). One such method, known as the CO_2 slicing technique, described in more detail in section 2, has been widely used to obtain the cloud top height and has been adapted for numerous instruments, as illustrated in table 2. The method has been shown to have some potential when applied to volcanic ash using the Moderate Resolution Imaging Spectroradiometer (MODIS) (Richards, 2006; Tupper et al., 2007). In this study, the technique has been adapted for the Infrared Atmospheric Sounding Interferometer (IASI; see section 3) and applied to volcanic ash. It was first applied to simulated ash spectra (section 4) to select the most appropriate channels and to demonstrate that the method has promise when applied to volcanic ash. Following this it was applied to scenes containing volcanic ash from the Eyjafjallajökull and Grimsvötn eruptions (section 5) where it was compared against an existing method for obtaining the height of volcanic ash and data from a satellite borne LiDAR. The results indicate that this method could be applied to get a first approximation of the ash cloud height which could then be used for hazard mitigation and as a parameter in other retrieval methods or ash models.

2 CO_2 Slicing

The CO_2 slicing technique is an established method, developed for obtaining the cloud top height/pressure of ~~meteorological~~ aqueous cloud (Chahine, 1974; Smith and Platt, 1978; Menzel et al., 1983). Over the past four decades this tool has been adapted for different instruments, summarised in table 2, including both airborne and satellite platforms. The technique uses a CO_2 absorption feature within the thermal infrared part of the electromagnetic spectrum between 665 and 750 cm^{-1} (13.3 to 15 μm). Within this region, as wavenumber increases there is a general increase in the radiance observed. This is demonstrated in Fig. 1a which shows the spectrum of a simulated clear atmosphere. This has been simulated with the fast radiative transfer model RTTOV (version 9; Saunders et al., 1998) and replicates what would be observed with IASI given specified atmospheric conditions. In this case a default atmospheric profile is used, without the addition of cloud, volcanic ash or any trace gases or aerosols above background levels.

~~Assuming an atmosphere which is decreasing in temperature~~ In the Earth's troposphere where temperature is decreasing with height, the radiances measured by the instrument are proportional to the transparency of the atmosphere for each channel (Holz et al., 2006). Subsequently, within the CO_2 absorption band, as wavenumber and the radiance measured both increase, the channels are becoming increasingly transparent (with some fluctuations). As such, the spectrum of a high altitude cloud will begin to deviate from the clear spectrum at a lower wavenumber than a lower altitude cloud. This is illustrated in Fig. 1a which ~~also~~ shows the spectra of three ash clouds of varying heights. Effectively, until the point where the clear and ash/cloudy spectra diverge, the instrument is recording clear radiances. This concept has been used to identify channels whose cloud free radiances can be assimilated into numerical weather prediction models, rather than filtering out these pixels entirely (e.g. McNally and Watts, 2003).

The changing sensitivity of each of the channels to the atmospheric profile is better demonstrated in Fig. 1b and c. This shows the derivative of atmospheric transmittance with log pressure ($d\tau/d\ln p$) and the peak of this value respectively. This is a measure of each channel's sensitivity to each level of the atmosphere and demonstrates that this shifts from the upper atmosphere at lower wavenumbers ~~to~~ towards the surface at higher wavenumbers.

As the channels are sensitive to different parts of the atmosphere it is possible to use this to estimate the height of the cloud (~~meteorological~~ aqueous or in principle ash). To do this using the CO_2 slicing method, the ratio (f, Eq. 1) of the difference in cloudy and clear radiances (L_{obs} and L_{clr} respectively) for two channels (ν_1 and ν_2) within or close to the CO_2 absorption band is compared against a cloud pressure function (C, Eq. 2):

$$f(\nu_1, \nu_2) = \frac{L_{obs}(\nu_1) - L_{clr}(\nu_1)}{L_{obs}(\nu_2) - L_{clr}(\nu_2)} \quad (1)$$

$$C(\nu_1,\nu_2,p) = \frac{N\varepsilon_1 \int_{p_s}^{p_c} \tau(\nu_1,p)\frac{dB[\nu_1,T(p)]}{dp}dp}{N\varepsilon_2 \int_{p_s}^{p_c} \tau(\nu_2,p)\frac{dB[\nu_2,T(p)]}{dp}dp} \qquad (2)$$

where τ is the atmospheric transmittance at channel ν ~~emitted at~~ of the layer between the pressure level p ~~arriving at the instrument~~ and the instrument (top of the atmosphere); B is the Planck radiance which is channel and temperature (and therefore pressure) ~~dependant~~dependent; p_c and p_s are the cloud and surface pressure respectively; and $N\varepsilon$ is the effective emissivity (sometimes referred to as the effective cloud amount), a product of the cloud fraction (N) and cloud emissivity (ε). Equation 1 is compared against Eq. 2 and where the two functions intersect is taken as the cloud top pressure. A demonstration of this is shown in Fig. 2a. Following this the effective emissivity can be computed using a channel which falls within an atmospheric window (w; usually one close to the CO_2 absorption band):

$$N\varepsilon = \frac{L_{\text{obs}}(w) - L_{\text{clr}}(w)}{B[w,T(p_c)] - L_{\text{clr}}(w)} \qquad (3)$$

In most applications of the CO_2 slicing technique, multiple channel pairs are used, resulting in different height solutions. In many studies, channel pairs are not considered if $L_{\sout{\text{cld}}\,\text{obs}}(\nu)$ - $L_{\text{clr}}(\nu)$ for either the CO_2 (ν_1) or reference (ν_2) channels used falls within the noise of the instrument at that channel (e.g. Menzel et al., 1992). The solution may also be rejected if the effective emissivity computed using Eq. 3 is not between 0 and 1.05 (e.g. Arriaga, 2007). If multiple solutions remain, then a number of different techniques can be employed to obtain a final value. This includes a top down approach where the solution of the most opaque channel is accepted if it is within an expected height range, and if not the next most opaque channel is considered. This is repeated until an appropriate height value is obtained (Menzel et al., 2008). Alternatively, the height and cloud fraction which best satisfies the radiative transfer equation for all the channels used is accepted as the final cloud pressure/height (e.g. Menzel et al., 1983, 1992). If all of the channel pairs are considered inappropriate, for example, if $L_{\sout{\text{cld}}\,\text{obs}}(\nu)$ - $L_{\text{clr}}(\nu)$ is within the noise of the instrument for all the channels used, then many methods assume that cloud is opaque and compare the brightness temperature measured by the instrument at 11 μm to an atmospheric temperature profile to obtain an alternative cloud height (e.g. Menzel et al., 1983, 1992; Zhang and Menzel, 2002; Menzel et al., 2008).

The issue of multiple solutions is further complicated for hyperspectral instruments as these can have hundreds of channels within the CO_2 absorption band. Some methods apply a weighting function based on each channel's sensitivity to the atmosphere (e.g Smith and Frey, 1990). However, to avoid a high computational cost, often there needs to be some prior consideration of the most appropriate channels. This has included exploring large datasets with known cloud top heights to select the most appropriate channels (e.g. Arriaga, 2007). Other approaches include the creation of synthetic channels by averaging the radiances of channels sensitive to the same portion of the atmosphere (Someya et al., 2016) or CO_2 sorting which looks for the point where the clear and cloudy spectra deviate which is the first point where the instrument can see the cloud layer (Holz et al., 2006).

The CO_2 slicing method makes a number of assumptions: (1) ~~That~~ the cloud is infinitesimally thin; (2) in cases where there are multiple layers of cloud, the lower level clouds are ignored; (3) the two channels used in Eq. ~~1~~ 1 are sufficiently close that the difference in emissivity between them is negligible; (3) in cases where there are multiple layers of cloud, the lower level clouds are ignored. The second : this is particularly important to consider when the channel pairs are selected. Multiple cloud layers have previously been identified as a source of error in the CO_2 slicing retrieval with the extent of this being affected by the channels used and the height of the underlying layers (Menzel et al., 1992). For example, an opaque cloud close to the surface is unlikely to affect the height retrieval of a cirrus cloud when using channels which are not sensitive to radiation from the lower troposphere. In contrast, a an opaque cloud in the middle of the troposphere might lead to the underestimation of the cloud top height of a higher cirrus layer (Menzel et al., 1992). The effect of surface emissivity is expected to be minimal as channels within the CO_2 absorption band have weighting functions which that peak above the surface, as shown in Fig. 1d.

An additional consideration has to be made when applying the CO_2 slicing method to volcanic ash. The height that a volcanic ash cloud reaches is largely dependant on the force of the eruption and the atmospheric conditions (Sparks et al., 1997) and so this can vary widely. Large explosive eruptions can generate columns which enter the stratosphere, which can then potentially affect climate (Robock, 2000). The cloud pressure function generated using Eq. 2 is temperature dependantdependent. Within the troposphere, the temperature decreases with height; however, in the stratosphere the temperature beings to climb again. This leads to a reversal in the cloud pressure function, which in some cases can result in multiple solutions: one in the troposphere and one in the stratosphere. Consequently, some prior information is required to determine whether the plume is within the troposphere and therefore if the CO_2 slicing technique is appropriate. This might include observations made on the ground or by pilots. The CO_2 slicing technique has previously only been used to determine the height of meteorological aqueous clouds in the troposphere and so in this study only the tropospheric solution is accepted.

3 The Infrared Atmospheric Sounding Interferometer

The Infrared Atmospheric Sounding Interferometer (IASI) is an instrument on-board three meteorological satellites, MetOp Metop A, B and C, launched in 2006, 2012 and 2018 respectively. Each instrument orbits the Earth twice a day. The instrument scans have a swath width of 2200 km and consist of groups of four circular pixels which have a diameter of 12 km at nadir (Clerbaux et al., 2009). The instruments measure across the infrared between 645 to 2760 cm^{-1} (3.62 to 15.5 μm) with a high spectral resolution of 0.5 cm^{-1} (Blumstein et al., 2004).

The instrument has previously been used to analyse volcanic plumes of SO_2 (e.g. Clarisse et al., 2008; Walker et al., 2012; Carboni et al., 2012; Clarisse et al., 2012, 2014; Carboni et al., 2016; Taylor et al., 2018) and ash (e.g. Clarisse et al., 2010; Maes et al., 2016; Ventress et al., 2016) from a number of different eruptions. Previous methods for determining the height of the plume with spectra measured by IASI use the optimal estimation method (Maes et al., 2016; Ventress et al., 2016). The CO_2 slicing method has previously been applied to IASI spectra to obtain the cloud top height of meteorological aqueous cloud (Arriaga, 2007). The values obtained for the cloud pressure and emissivity are often assimilated in numerical weather prediction models (Guidard et al., 2011; Lavanant et al., 2011). The different adaptations of the CO_2 slicing technique for IASI use different numbers and combinations of channels and can therefore give different results (Lavanant et al., 2011). In this study, channels are selected based on the technique's performance when applied to simulated ash spectra.

4 Application to simulated data

4.1 Channel selection

IASI has over 300 channels which fall within the CO_2 absorption band, and so, to ensure computational efficiency an appropriate subset of these channels must be selected. To do this the CO_2 slicing technique was first applied to 384 simulated ash spectra. These are 'ideal' test cases, which do not include other aerosols or aqueous cloud. These spectra include six different atmospheres: high latitude, mid-latitude day and night, tropical daytime and polar summer and winter . The simulated spectra also represent a range of different ash properties: (including atmospheric profiles created for MIPAS; Remedios et al. (2007)). The spectra were modelled using the refractive indices of samples of volcanic ash from the Eyjafjallajökull eruption in 2010 (Peter, 2010): the main eruption considered in this study. In the future different refractive indices could be used such as those in Prata et al. (2019). A range of ash properties were explored: cloud heights between 200 and 900 hPa (going slightly above the tropopause), ash effective radius between 5 and 10 μm, and ash optical depths ranging between 5 and 15 (referenced at 550 nm), ash effective radius between 5 and 10 . Typically, the effective radius is less than 8 μm for very fine ash (such as in a distal plume) and between 8 and 64 μm and cloud heights from 200 to 900 mbfor fine ash (Marzano et al., 2018). The range of ash optical depths is highly variable. Ventress et al. (2016) and Balis et al. (2016) recorded ash optical depths of less than 1.2 from dispersed plumes from Eyjafjallajökull in 2010; however much higher values can be expected closer to the volcano or following large explosive eruptions. The effective radius and AOD explored here for the channel selection is in the upper range and above what might be expected: values which may only be true close to the volcanic vent. The spectrum of an optically thin plume is more difficult to differentiate from a clear spectrum commonly leading to the signal $(I_{obs}(v) - I_{clr}(v))$ to be within the instrument noise and subsequently will result in no retrieval. A decision was made to select the channels used using idealised optically thick cases, which may only be true close to the vent, for which the plume should be evident in the majority of the CO_2 channels. The selected channels are tested on a wider range of AODs and effective radius, including smaller values that are more representative of a disperse plume, in section 4.2.

The CO_2 slicing method was first applied using every channel combination between 660 and 800 cm^{-1}, where the reference channel (ν_2) wavenumber is greater than the CO_2 channel (ν_1) wavenumber. In this way, the reference channel is generally more sensitive to a lower part of the atmosphere than the CO_2 channel. As with existing studies only tropospheric solutions were accepted and in cases where the curve of the cloud pressure function resulted in multiple solutions, the solution with the greater weight (in this case the weighting function is defined as $w = d\tau[\nu_1, p]/dlnp)$ $k = d\tau[\nu_1, p]/dlnp)$ was accepted. The output from each channel pair was only accepted if it met three quality control criteria: (1) $L_{\mathrm{obs}}(\nu_1)$- $L_{\mathrm{clr}}\nu(_1)$ must be greater than the noise of the instrument at channel ν_1 (CO_2 channel; within the CO_2 absorption band the noise of the IASI instruments is between 2.55×10^{-8} and 3.77×10^{-8} W/(cm^2.sr.cm^{-1}))(2) Similarly,$L_{\mathrm{obs}}(\nu_2)$ - $L_{\mathrm{clr}}(\nu_2)$ must be greater than the noise of the instrument at ν_2 (reference channel); (3) The solution to Eq. 3 must fall between 0 and 1.05 (following Arriaga 2007).

The results are shown in Fig. 3. The top two lines rows show the maximum pressure difference between the true (simulated) and CO_2 slicing retrieved values divided into each pressure level. In total there are 48 spectra for each pressure level with these incorporating the different atmospheric profiles and ash properties. The lower two lines rows of Fig. 3 show the percentage of accepted retrievals. This refers to where there was an intersection between the two functions shown in Eq. 1 and 2, and where the value retrieved meets all three quality control conditions. This is also grouped into the eight pressure levels. The equivalent plots for the six individual atmospheres can be seen in Fig. A1-A6 in the appendix. Potentially, the method used in this study to select the most appropriate channels, could be performed for the different atmospheres to select channels which might be more suited to specific climatologies.

Figure 3 demonstrates that the best performing ~~channels~~ channel pairs vary depending on the height of the plume. ~~As expected, this shifts from lower wavenumbers at lower pressures to higher wavenumbers closer~~ For plumes at lower pressures, the maximum pressure difference between the simulated and retrieved pressures is smaller at lower CO_2 wavenumbers. For example, for the plumes simulated at 300 hPa, the maximum pressure difference was lowest (less than 20 hPa) for CO_2 channels between 700 and 710 cm^{-1}. As the pressure of the ash layer is increased, values are longer obtained at smaller wavenumbers. For example, for a plume at 500 hPa, solutions are no longer obtained for CO_2 channels which are less than 700 cm^{-1}: the maximum pressure difference between the true and retrieved values is now smaller for slightly higher wavenumbers. For a plume at 800 hPa the maximum pressure difference is lowest (less than 60 hPa) for CO_2 channels between 715 and 720 cm^{-1}. This observation reflects what is shown in fig. 1b and c: that the channel's peak sensitivity shifts from higher in the atmosphere at lower wavenumbers to close to the surface as higher wavenumbers effecting the best performing channel combinations. Notably, at 200 ~~mb~~ hPa there are far ~~less~~ fewer channels which pass the quality control conditions, and where a retrieval is possible, there is a large difference between the true and retrieved pressure. It is also possible to identify an increased error closer to the surface. Previous studies have acknowledged that the CO_2 slicing tool is less successful at pressures greater than 700 ~~mb~~ hPa (Menzel et al., 2008) because approaching the surface there are ~~less~~ fewer channels with a distinction between the clear and cloudy spectra, often leading to $L_{\cancel{cld}obs}(v)$ - $L_{clr}(v)$ to be within the range of the instrument's noise and therefore the channels being excluded. Another observation that can be made from Fig. 3 is that channels below 700 cm^{-1} often have a low percentage of accepted retrievals. These channels are shown in Fig. 1b and c to be sensitive to the heights above the tropopause. This may also be the reason for few accepted retrievals at 720 cm^{-1}. Additionally, for channels greater than 750 cm^{-1}, which are no longer in the CO_2 absorption band, the difference between the true and retrieved pressure is usually greater than 100 ~~mb~~hPa.

Figure 4 shows a similar plot between 700 and 750 cm^{-1}. In this case, the spectra were also grouped into three categories: high cloud (300-400 ~~mb~~hPa), mid level cloud (500-600 ~~mb~~hPa) and low level cloud (700-800 ~~mb~~hPa). Note that the simulated spectra at 200 and 900 ~~mb~~ hPa have been excluded. Also, the maximum pressure difference is only shown where it is less than 75 ~~mb~~ hPa and where the percentage of successful retrievals is greater than 50%. This plot has been used to manually select the most appropriate set of channels. The best selection of channel pairs will be representative of the entire atmosphere (channels should be selected which peak at different heights~~(~~, Fig. 1c), while minimising the difference between the simulated and retrieved pressures, and maximising the acceptance rate, Fig. 4. Another consideration is the assumption that the change in emissivity between the channel pairs is negligible. The emissivity ratio for a sample of ash from the Eyjafjallajökull eruption (the main eruption considered in this study) for all channel combinations in the 680 and 800 cm^{-1} range is shown in Fig. 5. For this assumption to hold true, the emissivity ratio should be as close to 1 as possible. This is usually the case for channels which are close together. Given these criteria appropriate channel ranges have been selected. These channel ranges and the reference channels are shown in table 3. The weighting functions for the selected channels are shown in Fig. 1d.

4.2 Simulation results

Following the selection of channels, the final pressure values (P) were computed by taking a weighted average of the results:

$$P = \frac{\sum p_c(\nu)k^2(\nu)}{\sum k^2(\nu)} \tag{4}$$

where p_c is the pressure retrieved for channels ν and ~~w~~ k refers to the weighting function based on the derivative of atmospheric transmittance computed for each pressure level with RTTOV with respect to the log of atmospheric pressure ($d\tau[\nu,p]/dlnp$). On this occasion, the retrieval was applied to 1344 simulated ash spectra including those with lower ash optical depths (ranging from 0.5 to 15) and smaller effective radius (ranging from 1 to 10 μm). This includes spectra representative of thinner ash clouds which were not considered during the channel selection.

The results are displayed in Fig. 6a-f which plots the true (simulated) pressures against the final weighted pressures obtained with the CO_2 slicing technique. The different atmospheres are displayed separately and the percentage of accepted retrievals are indicated below each plot. Table 4 reports the root mean square error (RMSE) for each atmosphere. Overall, the CO_2 slicing method returned values for 72% of the simulated spectra, with an RMSE of 777 m. These results suggest that the technique does have merit for obtaining the height of ash clouds.

Figures 6g-i give some indication of where and why the retrieval was unsuccessful. Figure 6g-h show there are slightly more failed cases for ash spectra with the lowest optical depth (0.5) and effective radius (1 μm). These low values are representative of thinner ash clouds whose spectra are more similar to clear atmospheric spectra. Subsequently, these cases are likely to fail the signal/noise quality control tests (Menzel et al., 1992, 2008). For example, an ash cloud at 500 ~~mb~~ hPa only has 7 channels which pass the $L_{\text{obs}}(\nu_1)$ - $L_{\text{clr}}(\nu_1)$ quality control condition when the ash optical depth is 0.1. However, the number of channels passing this criterion increases to 38 at an ash optical depth of 2.3. This observation is supported by Fig. 6j-k which shows the number of cases where the difference between the simulated and retrieved pressure is less than 0.5 km: which is slightly lower for a smaller effective radius and ash optical depth.

The majority of failed cases are shown to be at the pressure extremes, Fig. 6i. Similarly, Fig. 6l indicates that there are fewer cases where the pressure difference between the simulated and retrieved pressures are less than 0.5 km at these pressures. Close to the surface this can again be attributed to less distinction between the clear and ashy spectra (Menzel et al., 2008). For example, for the RTTOV default atmosphere, an ash plume at 900 ~~mb~~ hPa fails the signal/noise condition for all the channels used regardless of the optical depth and effective radius of the simulation. The lowest simulation pressure (200 ~~mb~~hPa) is close to or above the tropopause for all six atmospheres and for this example the CO_2 slicing method was allowed to retrieve up to the height of the reversal of the temperature profile (which is slightly above the tropopause). At these heights, the temperature gradient (dT/dp) is relatively stable, causing a similar effect in the cloud pressure function (best illustrated in Fig. 2) and subsequently a greater number of unsuccessful retrievals: the CO_2 slicing technique has previously been shown to perform poorly in isothermal regions of the atmosphere (Richards et al., 2006). This may also be the reason for the poor performance of the CO_2 slicing technique when applied to the polar summer atmosphere for which the technique only retrieved values for 29% of cases.

The RMSE and the percentage of accepted retrievals for the CO_2 slicing technique, without the quality control criteria applied, are shown in table 4. Figure A7 shows the equivalent plot to Fig. 6 without the quality control. The addition of the quality control compromises the number of successful retrievals for an overall reduction in the RMSE. Overall, the reduction is around 200 m but in individual cases by up to 1.4 km (e.g. tropical atmosphere). Figure A7 indicates that the addition of the quality control is particularly advantageous for lower level ash layers which without the quality control are often overestimated. Overall, the results show that this adaptation of the CO_2 slicing technique has promise for obtaining the height of volcanic ash clouds within the troposphere, although its use is limited in cases of low level or thin clouds or where there is a steep temperature gradient.

5 Application to scenes containing volcanic ash

The CO_2 slicing method has been applied to scenes containing ash from the Eyjafjallajökull (63.63°N, 19.63°W, 1651 m) and Grimsvötn (64.42°N, 17.33°W, 1725 m) eruptions in 2010 and 2011 respectively. The plumes from both eruptions were closely monitored using a variety of instrumentation which included ground based remote sensing, airborne measurements and the use of satellite products (e.g. Gudmundsson et al., 2010; Weber et al., 2012). The ash and gas clouds from these eruptions have since been extensively studied (e.g. Kerminen et al., 2011; Tesche et al., 2012; Flemming and Inness, 2013; Cooke et al., 2014; Ventress et al., 2016). They are commonly used to demonstrate the utility of new remote sensing developments (e.g. Mackie and Watson, 2014; Taylor et al., 2015; Ventress et al., 2016; Western et al., 2017), and similarly are often used in modelling research (Matthias et al., 2012; Webster et al., 2012; Moxnes et al., 2014; Wilkins et al., 2016). This makes them the ideal first candidates for the CO_2 slicing technique. Another reason for choosing these eruptions is that in both cases, the ash clouds were confined to the troposphere making them an appropriate target for the CO_2 slicing technique.

In this application of the retrieval, it has only been applied to pixels which are flagged as containing volcanic ash by a linear ash retrieval developed for IASI (~~Ventress et al. 2016~~Ventress et al. 2016; Sears et al. 2013: following the method developed for SO_2 by Walker et al. 2012). ~~Values~~ This method compares each IASI spectra against a covariance matrix formed from pixels which contain no volcanic ash thereby representing the spectral variability associated with interfering gas species or clouds, and also the instrument noise. A least squares fit is performed for three ash altitudes (400, 600 and 800 hPa) to retrieve a value for ash optical depth. A pixel is then flagged if it exceeds a threshold at any height. As SO_2 can, with caution, be used as a proxy for volcanic ash (Carn et al., 2009; Thomas and Prata, 2011) the retrieval has also been run for pixels flagged for SO_2 using the same approach (Walker et al., 2011, 2012; Carboni et al., 2012, 2016). For the CO_2 slicing values for L_{clr} were obtained using the radiative transfer model RTTOV using the ECMWF atmospheric profile as an input and using the default ocean emissivity within RTTOV. The effect of surface emissivity is thought to be minimal as for the channels used the weighting functions peak above the surface, Fig. 1d. The temperature and humidity profiles needed to calculate the ~~planck~~ Planck radiance and τ were acquired from the European Centre for Medium-Range Weather Forecasts (ECMWF). The closest ECMWF profile to each individual IASI pixel was used. RTTOV was used to compute the transmittance values. Another point to note is that, in section 4, the maximum height that could be retrieved was defined as the height at which the temperature profile inverts and has a positive gradient. Figure 6 demonstrated that the CO_2 slicing method performs poorly where the temperature profile steepens significantly. For the application to real satellite data, the maximum height which can be retrieved is the height of the tropopause as This is slightly above the tropopause which is defined by the World Meteorological Organisation : (WMO) as the point at which the lapse rate is less than 2°C/km, and remains lower than this for at least 2 km. This was done to demonstrate how the CO_2 slicing method performs above the troposphere where the atmospheric temperature does not vary significantly: the atmospheric lapse rate here approaches zero. Figure 6 demonstrates that the CO_2 slicing method performs poorly in these cases and so in the application to real data the CO_2 slicing method is only able to retrieve values up to the tropopause as defined by the WMO.

5.1 Methods used for comparison

5.1.1 Optimal Estimation Scheme

The CO_2 slicing plume altitude results have been compared against the plume altitude obtained using the optimal estimation (OE) retrieval scheme developed by Ventress et al. (2016). The retrieval scheme combines a clear-sky forward model with a (geometrically) infinitely thin ash layer to simulate atmospheric spectra, using ECMWF data as input atmospheric parameters. The simulated spectra are compared to the satellite measurements and, using the cost function (a measure of retrieval fit), the spectrum that most closely matches the spectrum obtained with IASI is used to determine the ash plume properties. This method retrieves the effective radius and ash optical depth, which can be used to calculate the mass of ash within the plume. For more information on this technique, refer to Ventress et al. (2016).

5.1.2 CALIOP

While a comparison against another IASI retrieval is useful, such comparisons have limitations. All retrieval techniques make assumptions and have different limitations and so it is not expected that the results would be the same, or even similar, in all cases. An additional comparison is made with the Cloud-Aerosol LiDAR with Orthogonal Polarization (CALIOP) instrument, on-board the the Cloud-Aerosol LiDAR and Infrared Pathfinder Satellite Observations (CALIPSO) satellite. This active sensor was launched in 2006 and forms part of NASA's afternoon constellation (A-Train) of satellites. The instrument has a 30 m vertical resolution and 335 m spatial resolution, and orbits roughly every 16 days (Winker et al., 2009; Hunt et al., 2009). The backscatter profile obtained with LiDAR instruments can be used to obtain the vertical structure of the atmosphere, providing information on the height and thickness of different scattering layers, including both ash and cloud. CALIOP and other LiDAR instruments are commonly used as a tool for the validation of cloud heights, including previous studies with the CO_2 slicing technique (e.g. Smith and Platt, 1978; Frey et al., 1999; Holz et al., 2006, 2008), and a number of ash retrievals (e.g. Stohl et al., 2011; Ventress et al., 2016).

To conduct a comparison between the heights obtained using the CO_2 slicing and OE techniques with CALIOP the data from the two instruments was first collocated. CALIOP overpasses which intersected with the ash plumes were identified using false colour images from the Spinning Enhanced Visible and Infrared Imager (SEVIRI) (Thomas and Siddans, 2015). The backscatter profiles were then averaged vertically to a 250 m resolution. The CALIOP data was smoothed to IASI's spatial resolution of 12 km and collocation was identified where measurements made by the two instruments fell within 50 km and 2 hours of each other. If multiple CALIOP pixels were matched to an IASI pixel then the CALIOP pixel which was closest in distance was selected for comparison. A cloud top height is obtained from the backscatter profiles allowing a comparison with the CO_2 slicing and OE methods. This was done by (1) calculating the mean backscatter above 15 km and subtracting this from the total backscatter; (2) for each pixel a cumulative backscatter is calculated; (3) the cloud altitude is where the atmospheric extinction exceeds a specified threshold. This threshold has been manually set for each scene, chosen to obtain the best match to the cloud top height shown in the CALIOP backscatter images.

5.2 Comparison of results

The CO_2 slicing technique was applied to IASI ash flagged pixels from 13 and ~~4~~ 3 days from the Eyjafjallajökull and Grimsvötn eruptions respectively. Maps of these results, with the orbits divided into morning and afternoon are shown in Fig. 7. For each map there is a histogram showing the distribution of the retrieved heights. Encouragingly, initial examination of the maps shows that the retrieved values are spatially consistent with only a few outliers. These outliers are usually individual pixels whose altitudes are higher than those surrounding them. Below each map are numbers indicating the total number of pixels in each plot and the number of pixels for which the CO_2 slicing technique was unable to obtain a height, either because there is no intersection between the two functions shown in Eq. 1 and 2 or because of the failure of one or more of the quality control measures outlined in section 4. Overall, the CO_2 slicing technique was able to obtain a height value for 88% of pixels from the two eruptions.

The CO_2 slicing results have been compared against those obtained with an optimal estimation (OE) scheme. Distributions of the heights obtained for all pixels from the two eruptions are shown in Fig. 8a and b. In both cases, the peak of the distribution for the CO_2 slicing heights is higher than for the OE scheme. Figure 9 shows how the average height obtained with the two retrievals has changed over the 13 days studied from the Eyjafjallajökull eruption. This plot shows that on the 5^{th} May the CO_2 slicing method retrieved an average altitude of roughly 7 km and that this then fell throughout the remainder of the study period. This corresponds to observations made about the volcano's activity. Activity at the volcano became more explosive on the 5^{th} May 2010 with increased emission of ash and SO_2, with plumes rising to greater than 8 km. This was followed by a fall in the plume height to 6-7 km: interspersed with higher plumes during more explosive activity (Petersen, 2010). The average CO_2 slicing heights shown in Fig. 9 are probably lower because these are values for the entire plumes including further away from the source. However, it does capture the changing elevation of the plume throughout the eruption. By contrast, the OE average heights are less variable: between 3 and 4.25 km throughout the period studied. Some example maps of the OE ~~results~~ heights are shown in Fig. 10 to 13b, alongside the ash mass (panel c) calculated from the OE retrievals of AOD and effective radius, assuming an ash density. The maps of ash mass show that in general the ash mass falls with transportation away from the vent: the plumes become more disperse. The different design, assumptions and limitations of the two techniques mean that it is not expected that the two retrievals will return the same or even similar values. The persistently lower average height for the OE technique suggests that it is strongly influenced by the height a priori (which was optimal estimation scheme uses only 105 channels between 680.75 and 1204.5 cm^{-1} ($\sim$ 8.3 - 14.6 μm) to improve computational efficiency. This includes 14 channels within the CO_2 absorption band, only one of which is in common with the CO_2 slicing. However, unlike the CO_2 slicing method presented here, the channels used by the optimal estimation scheme have not been optimised for retrieving the height of the ash layer. Ventress et al. (2016) noted that the optimal estimation retrieval could be further refined by altering the channels used. For example, channels with more height information could be selected. Similarly, Ventress et al. (2016) suggested that channels could be selected to minimise the effect of the underlying cloud layers following observations that the OE method can underestimate the cloud top height in cases of multiple cloud layers (Ventress et al., 2016). In the current application of the optimal estimation scheme, where there is not sufficient information about the height of the ash layer within the channels used, the retrieval height output will tend to the a priori height which in this case is around 3.5 km). This is potentially the reason for the persistently lower average height shown in Fig. 9 which suggests a strong dependence on the a priori. In future applications of the OE scheme, the CO_2 slicing results could be used as the a prioipriori if the one CO_2 channel that the two retrievals have in common was removed from the optimal estimation scheme. Other differences in the results may arise from the nature of the two techniques. The OE scheme returns values for the ash optical depth, effective radius and height by fitting simulated spectra to those obtained with IASI. Ventress et al. (2016) identified that in some cases the retrieval assumed a lower altitude and a higher underestimated the altitude of the plume and obtained a high ash optical depth in order to fit the spectra. Additional differences may arise from the channels used. As explained in section 4, the channels used for the CO_2 slicing have been specifically chosen for their ability to obtain the ash cloud height of simulated data. In contrast, Ventress et al. (2016) suggested that the OE height retrieval could be further refined by altering the channels used. One suggestion was to select channels which minimise the effect of the underlying cloud layers following observations that the OE method can underestimate the cloud top height in cases of multiple cloud layers (Ventress et al., 2016)measured spectra when in reality the ash layer might have a lower optical depth and higher altitude.

A comparison has been made against backscatter profiles and cloud altitudes obtained with CALIOP, to assess how successfully the two retrievals perform. These backscatter profiles are shown in Fig. 10-13d. The heights obtained from the OE and CO_2 slicing methods for pixels which fall within 2 hours and 50 km are overplotted, along with the heights obtained with CALIOP and the tropopause height. In these plots it is possible to observe that both methods are capable of capturing the height of the ash layer, but there are clear cases where one technique outperforms the other. In Fig. 10 which shows the backscatter plot for the 6^{th} May 2010, the CO_2 slicing method places the ash cloud between 5 and 7 km between 57.5 and 60.5°N. This is shown to be higher than the CALIOP heights (4-5 km) to which the OE results are a closer match. In the same image, between 63 and 64°N the CO_2 slicing results are again higher than the OE results but this time are closer to, but lower than, the heights obtained from CALIOP. The lower heights of both the CO_2 slicing and OE scheme relative to CALIOP might be related to the thick underlying cloud layer. Figure 11d shows another example from the 9^{th} May 2010. Here between 51 and 53°N the heights obtained with both methods match those obtained with CALIOP. However, further north between 56 and 60°N, the

CO_2 slicing results agree more closely compared to those from the OE scheme. At 66°N, the CO_2 technique obtains a value close to the cloud top height, whereas the OE scheme obtains a value which is more representative of a lower layer of cloud. Figures 12 and 13 shows examples from the Grimsvötn eruption and in both cases both height retrievals are shown to resemble the shape of the ash cloud layer shown by CALIOP. There are cases where both retrievals underestimate the cloud top height which may be due to multiple layers of cloud.

Pearson's correlation values and the root square mean error (RMSE) were computed to compare the two retrieval methods against the heights obtained with CALIOP. These are shown in table 5 and scatter plots comparing the retrieved values are shown in Fig. 8c and d. The Pearson's correlation values are greater for the CO_2 slicing than for the OE scheme, while the RMSE values are lower: 2.2 and 2.1 km for the the Eyjafjallajökull and Grimsvötn eruptions respectively for the CO_2 slicing technique, compared to 3.2 and 2.4 km obtained for the OE method. This implies an improved height retrieval from the CO_2 slicing method.

Although comparisons against LiDAR backscatter profiles are a common way of validating retrievals of ash and ~~meteorological~~ aqueous cloud height, these comparisons can be limited. CALIOP and IASI measure different things. The first measures backscattering while the latter measures thermal emission. Measurements are made with significantly different spatial resolutions (335 m compared to 12 km for CALIOP and IASI respectively) and in different locations (a maximum difference of 50 km). Clouds can also vary significantly in very short spaces of time. Although only pixels with a difference of 2 hours have been considered in this comparison, this is still sufficient time for changes in the cloud's position both vertically and horizontally. These may account for some of the differences seen between the CALIOP profiles and the results obtained with the CO_2 slicing and the OE scheme. The cloud heights obtained from the CALIOP profile are not always a perfect representation of the cloud top height which may also contribute to the differences observed. Although these limitations exist, comparisons against LiDAR instruments are still one of the best methods for validating cloud heights, and in this case demonstrate that the CO_2 slicing technique has potential as a tool for obtaining the cloud top height of volcanic ash.

6 Conclusions

The CO_2 slicing technique is an established method, used for decades, for retrieving the cloud top height of ~~meteorological~~ aqueous cloud. Although it has previously been acknowledged that it can be applied to volcanic ash, it is not commonly used for this purpose, and it has only been applied to MODIS. In this study, the technique was adapted for IASI using simulated ash data to select the most appropriate channels and then demonstrate the technique's capability. When applied to the simulated data, the technique was shown to perform well in five out of six atmospheres. However, an increased failure rate, was seen above and close to the tropopause and close to the surface. This was also true of ash with lower optical depths and effective radius. Similar observations have been made by previous CO_2 slicing studies. In this application three quality control criteria have been applied which successfully remove the majority of cases where there are large differences between the true and retrieved pressures. When applied to ash scenes from the Eyjafjallajökull and Grimsvötn eruptions, the CO_2 slicing results compared well against the CALIOP backscatter profiles. It was also demonstrated that the CO_2 slicing method obtained heights which more closely matched CALIOP than the optimal ~~estimimation~~ estimation scheme used for comparison.

This is the first application of the CO_2 slicing technique to obtain the height of volcanic ash from IASI spectra, and the results are very encouraging. One advantage of this algorithm is that it can be run fairly quickly and so it could be applied to get a first approximation of the height, which could then be used to help assist hazard mitigation. It can also then be used as an input parameter into models of ash cloud propagation or as an a priori in other retrieval schemes. There is also potential for the further development of this technique in the future. Previous applications to cloud have created synthetic channels (multiple channels averaged together) which could be used to further improve the algorithm and its sensitivity to lower level clouds (Someya et al., 2016). It would also be possible to explore other options for selecting channels or obtaining the final cloud height. The channel selection in this study was based on simulated data in six different atmospheres, another avenue to explore would be the selection of atmospheric specific channel pairs. Further work would also help appreciate the strengths and limitations of this technique, and therefore where its use is most appropriate.

Data availability. The data used in this paper can be made available by contacting the author (isabelle.taylor@earth.ox.ac.uk)

Appendix A

Some additional figures are included within this appendix. Figures A1 to A6 show the maximum difference between the true (simulated) and retrieved pressures for the six investigated atmospheres for all the channel combinations between 660 and 800 cm^{-1}. The plots are divided into the different pressure levels. The figure also includes the percentage of successful retrievals (where there is an intersections between the two functions shown in Eq. 1 and 2 and all quality control conditions are met). This is out of a total of 8 simulations (for each pressure level) with ash optical depths ranging between 5 and 15, effective particle radius ranging between 5 and 10 μm. These could be used to select channels which are appropriate for specific climatologies. Figure A7 shows the final simulation result for each atmosphere without the quality control applied.

Competing interests. There are no competing interests at present

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Five of the atmospheric profiles used for the channel selection are reference spectra for the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS; Remedios et al. 2007). The IASI spectra used in this study are available from the Centre for Environmental Data

Analysis (EUMETSAT, 2009). Atmospheric profiles needed to run the CO_2 slicing technique were obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF).

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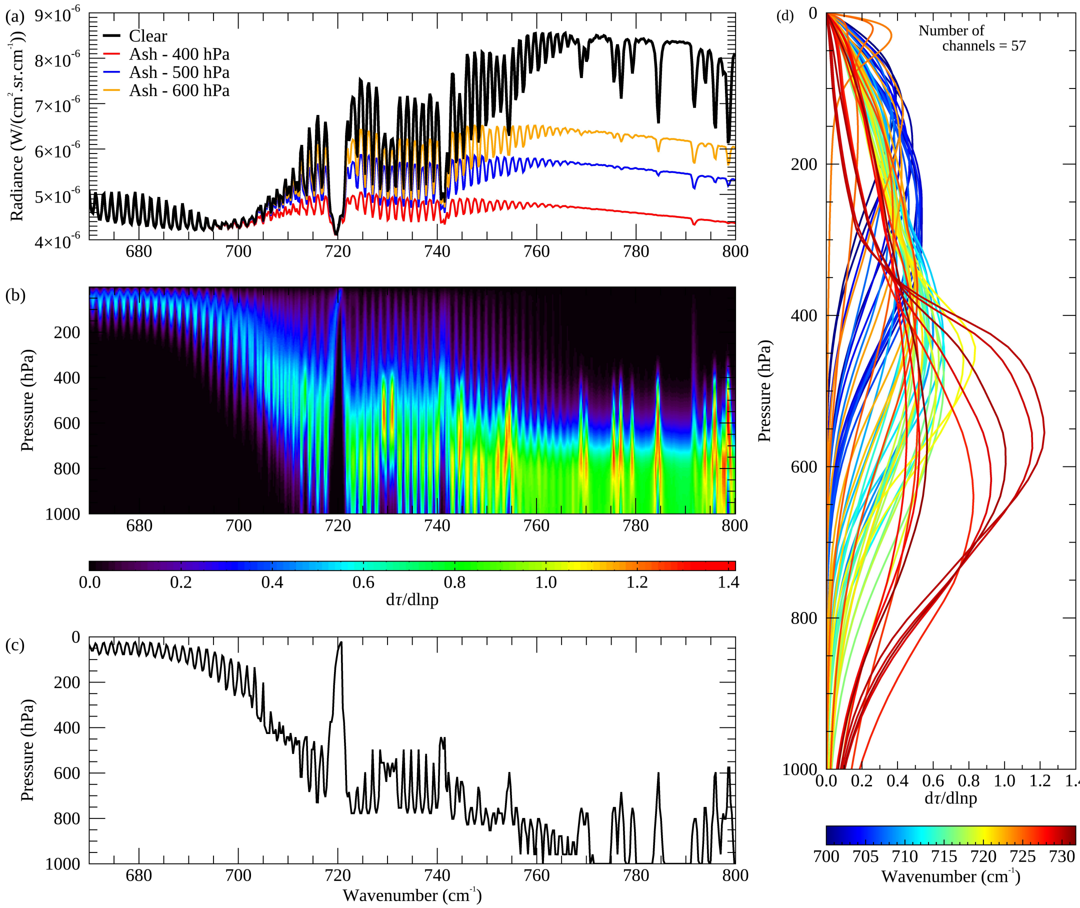

Figure 1. (a) Simulated spectra for a clear atmosphere (i.e. one without cloud or ash) and three ash clouds at different pressure levels: 400, 500 and 600 ~~mb~~hPa. (b) The change in atmospheric transmittance with log pressure ($d\tau/dlnp$). This is indicative of which part of the atmosphere each channel is sensitive to. This sensitivity is shown to shift from higher up in the atmosphere to the lower parts of the atmosphere as wavenumber increases. (c) The peak sensitivity for each channel. (d) The weighting function ($d\tau/dlnp$) for the 57 channels used in this CO_2 slicing study.

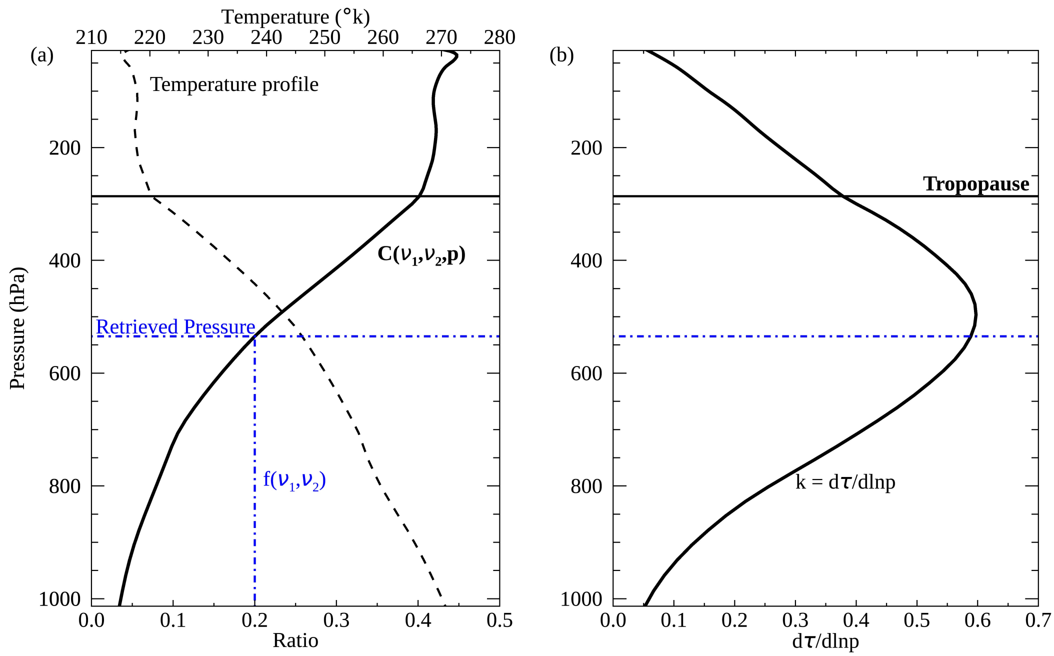

Figure 2. (a) An example of the cloud pressure function calculated using Eq. 2. This is strongly linked to the atmospheric temperature profile (dashed black line). The value obtained with Eq. 1 is compared against the cloud pressure function and where these intersect is taken as the cloud pressure solution for that channel. In this example ν_1 and ν_2 are at 715 cm^{-1} and 725 cm^{-1} respectively. (b) The corresponding weighting functions ($d\tau/dlnp$) which illustrates the changing sensitivity to the atmosphere. This is used to obtain a weighted average from multiple channel solutions.

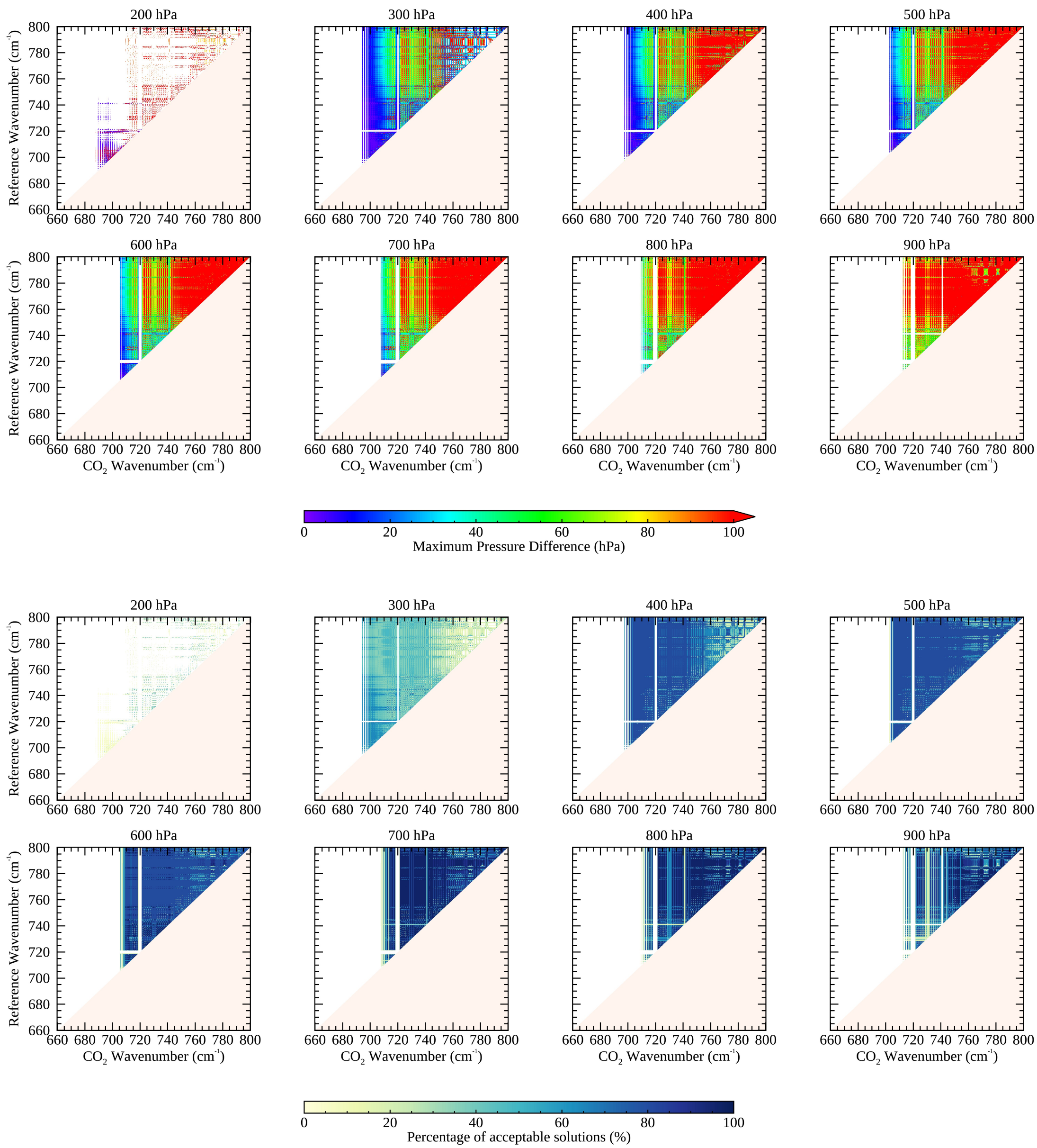

Figure 3. CO_2 slicing results for simulated ash spectra. The technique has been applied for each channel pair between 660 and 800 cm^{-1}. A total of 384 spectra were used which includes six different atmospheres. It also includes ash optical depths between 5 and 15, effective radius ranging between 5 and 10 μm and pressures between 200 and 900 ~~mb~~hPa. The first two ~~lines~~ rows of the plot show the maximum difference between the known (simulated) pressure and the pressure retrieved with the CO_2 slicing algorithm. This is divided into each pressure level. The last two ~~lines~~ rows show the percentage of successful retrievals. This is again divided into the 8 different pressure levels. In these plots the colour white indicates where no successful retrieval has been made and off white indicates channel combinations not explored in this study.

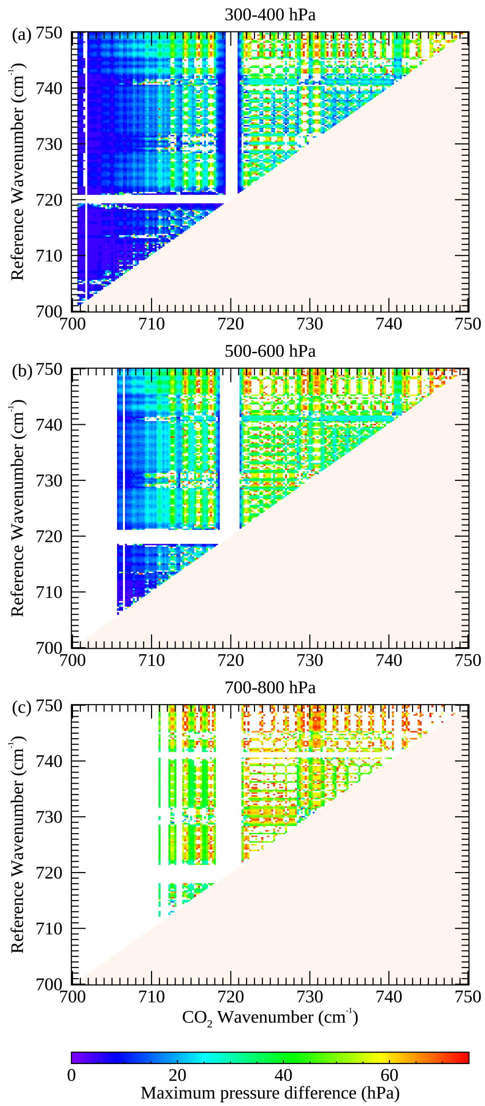

Figure 4. CO_2 slicing results for RTTOV simulated ash spectra. The plots show the maximum difference between the true (simulated) pressure and the pressure obtained with the CO_2 slicing algorithm. The results are split into three pressure levels: (a) high cloud (300-400 ~~mb~~hPa), (b) mid level cloud (500-600 ~~mb~~hPa) and (c) low level cloud (700-800 ~~mb~~hPa). Note that in this plot, results for 200 and 900 ~~mb~~ hPa are excluded. Results are only included where the maximum difference is less than 75 ~~mb~~ hPa and the percentage of successful retrievals is greater than 50%. This ~~is~~ was used to inform the choice of channels for the final CO_2 slicing algorithm.

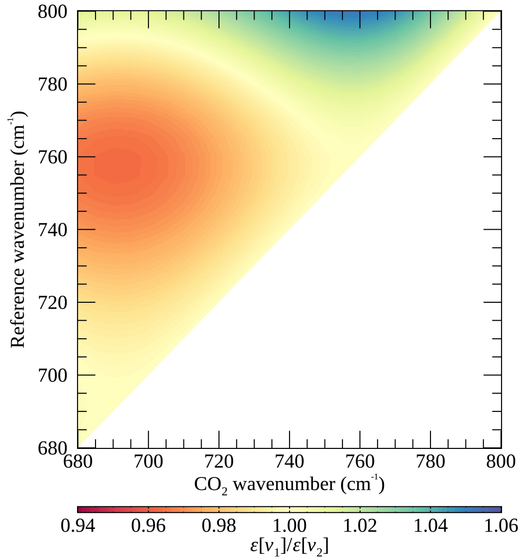

Figure 5. Emissivity ratio for channels between 680 and 800 cm^{-1}. The ash sample was from the Eyjafjallajökull eruption in 2010. The assumption that the emissivity does not vary significantly for the pair of channels used for the CO_2 slicing is important. For this to hold true, ideally the emissivity ratio should be close to 1.

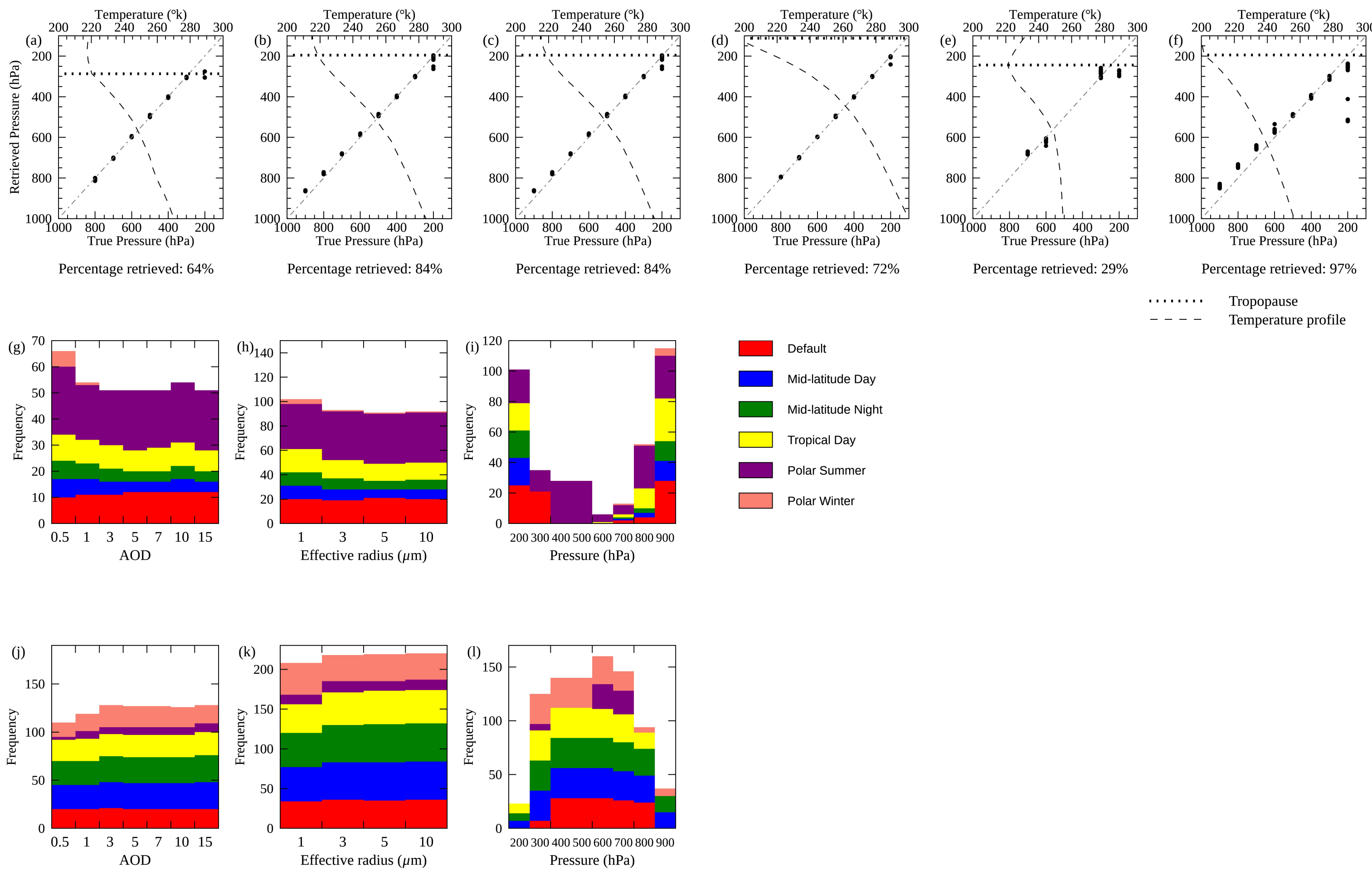

Figure 6. Final CO_2 slicing pressure results for RTTOV simulated ash spectra (a total of 224 spectra per atmosphere). The plots Panels (a)-(f) show the true (simulated) pressure plotted against the CO_2 slicing retrieved value for the six different atmospheres. (a) RTTOV default atmosphere (high latitude), (b) Mid-latitude day, (c) Mid-latitude night, (d) Tropical day, (e) Polar summer (f) Polar winter. In this case, the simulated spectra include the following ash properties: ash optical depth ranging between 0.5 and 15, ash effective radius ranging between 1 and 10 μm and pressure values between 200 and 900 mbhPa. Below each plot is a value indicating the percentage of successful retrievals (where a height value can be obtained and all quality control conditions have been met). (g) the The frequency of ash optical depths for which the CO_2 slicing technique was unable to return a height value. (h) Same as (g) for the effective radius. (i) Same as (g) for the ash cloud pressure. (j) The frequency of ash optical depths for which the difference between the simulation and CO_2 slicing height is less than 0.5 km. (k) The same as (j) for effective radius. (l) The same as (j) for ash cloud pressure. Related statistics can be seen in table 4. The equivalent plot, where the values which have not met the quality control conditions was not applied has been included in the appendix, figure A7.

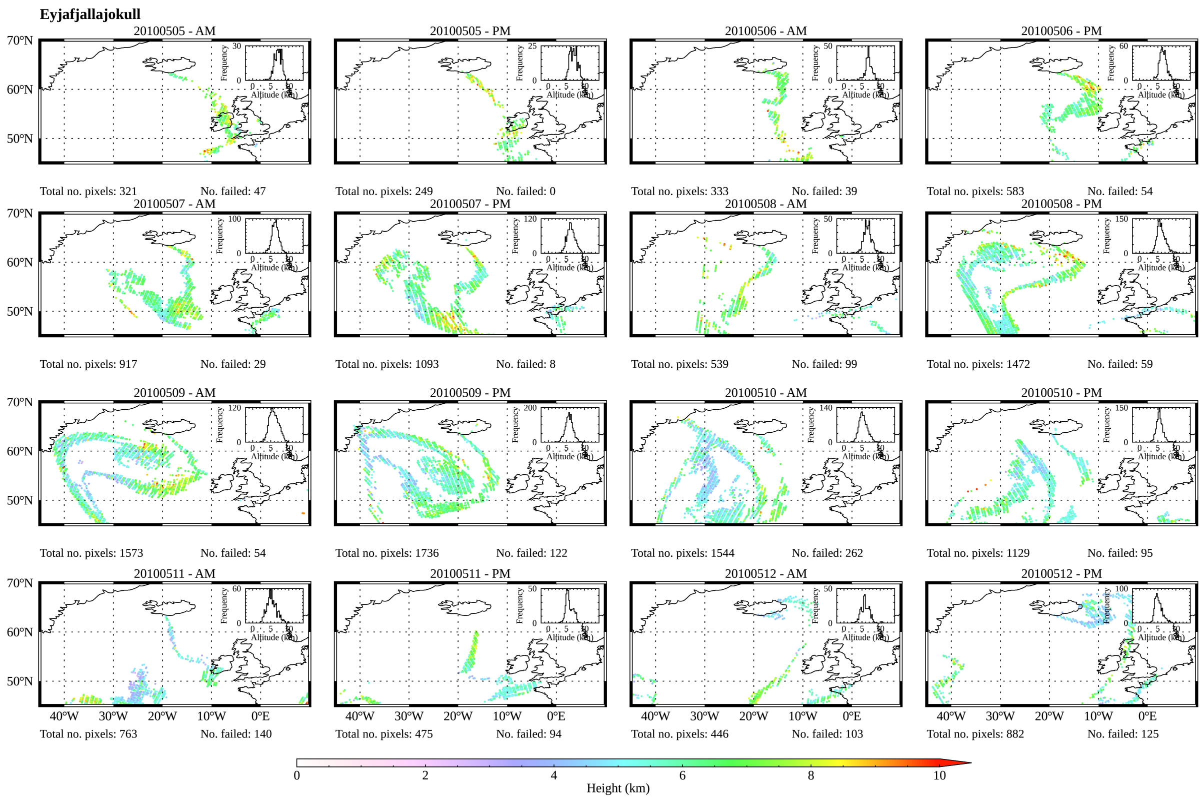

Figure 7. Maps of the CO_2 slicing output (with quality control applied) for the Eyjafjallajökull and Grimsvötn eruptions. Each plot consists of multiple orbits, divided into morning and afternoon. On each plot is a histogram showing the distribution of heights for each scene. Beneath each plot are numbers showing the total number of pixels in each image and the number of pixels for which the CO_2 slicing method was unable to return a value.

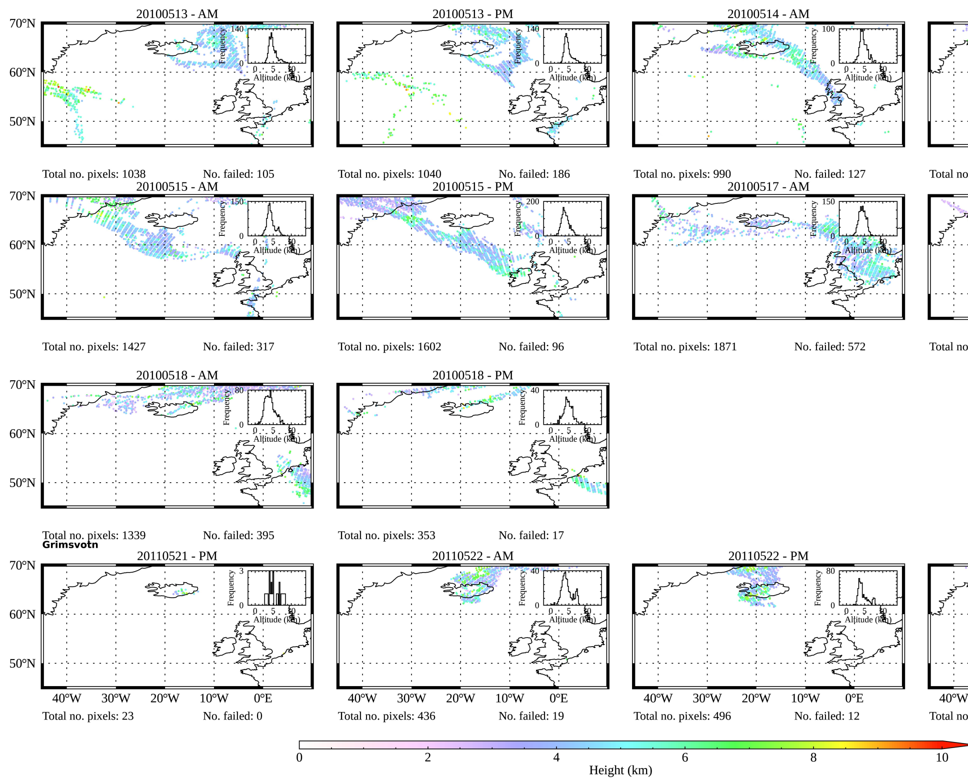

Figure 7. *continued*

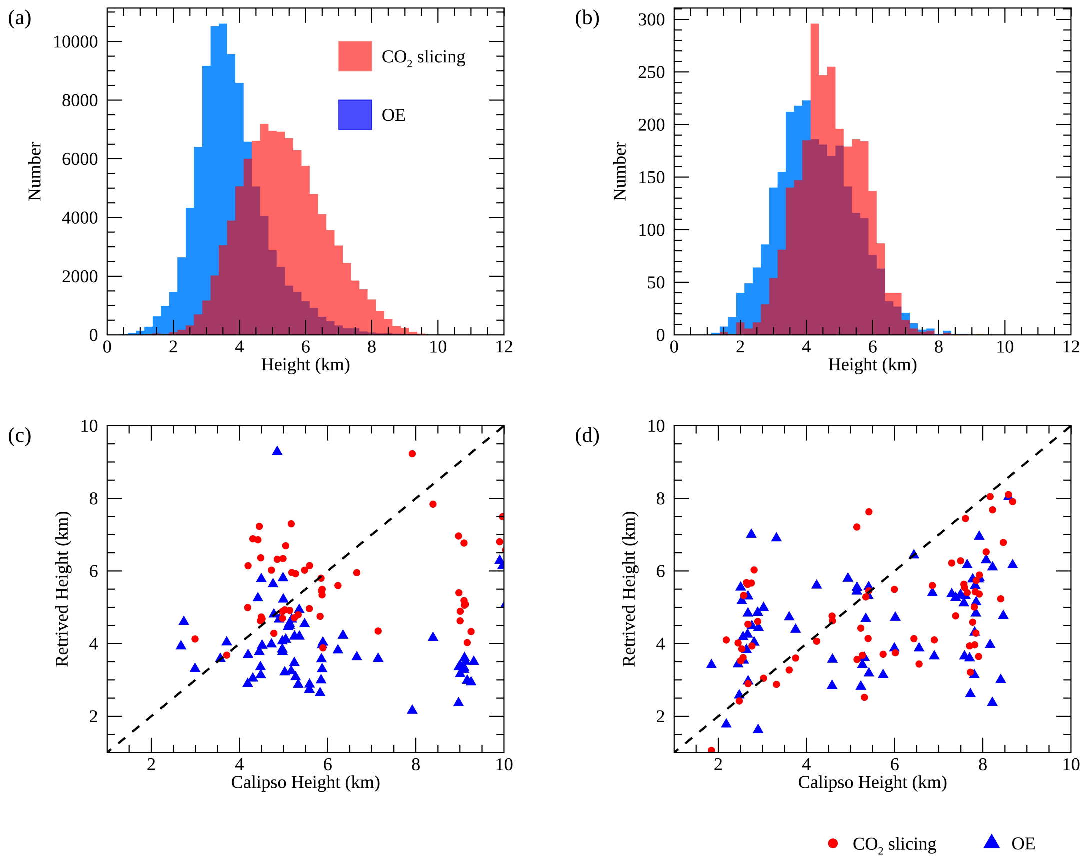

Figure 8. (a) Distribution of the CO_2 slicing and optimal estimation retrieved ash heights for all pixels from the Eyjafjallajökull eruption. (b) ~~Distribution of the retrieved ash heights~~ Same as (a) for the Grimsvötn eruption. (c) Comparison of the CALIOP heights with those obtained with the CO_2 slicing and optimal estimation techniques for a subset of pixels (where measurements fell within 50 km and 2 hours of each other) from the Eyjafjallajökull eruption. (d) Same as (c) for the Grimsvötn eruption. Related statistics can be seen in table 5.

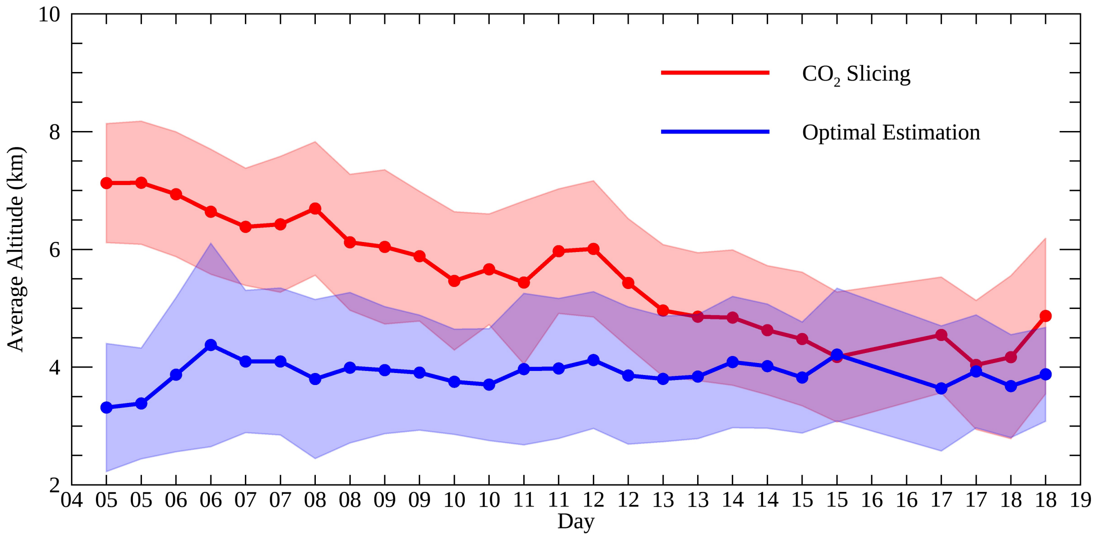

Figure 9. Time series showing how the average retrieved height for the CO_2 slicing and optimal estimation techniques varies during the Eyjafjallajökull eruption. The shaded polygon represents one standard deviation from the mean.

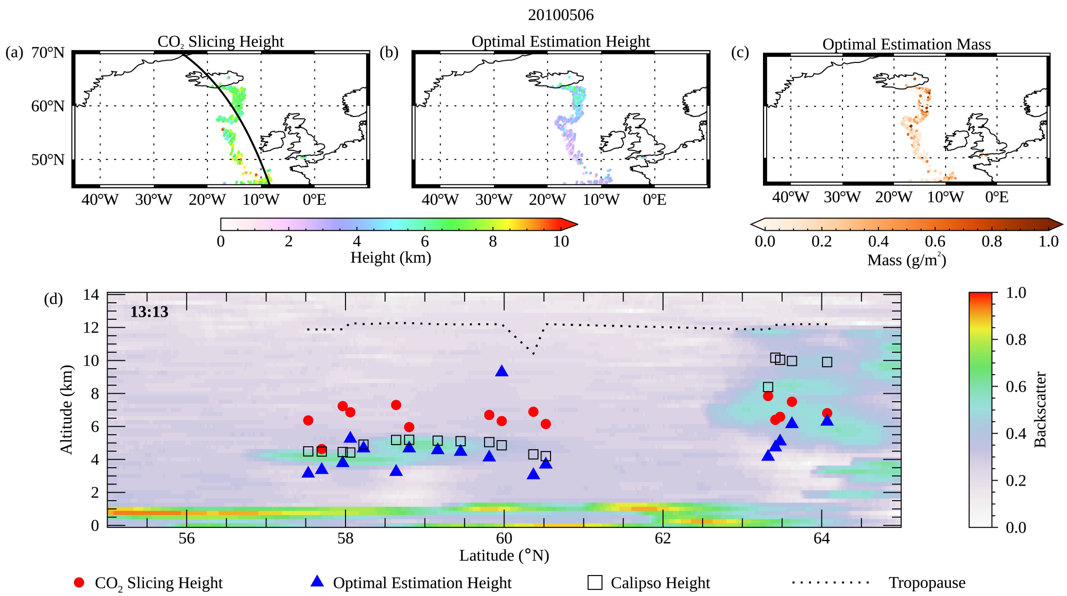

Figure 10. (a) CO_2 slicing results for the ~~6th~~ 6^{th} May 2010. Overplotted on this is the CALIPSO track. (b) The optimal estimation scheme heights. (c) The ash mass obtained with the optimal estimation scheme. (d) The CALIOP backscatter plot, with the CO_2 slicing results and the optimal estimation scheme heights plotted on top. Indicated on the top left hand side of the plot is the time of the CALIOP overpass. The dashed line indicates the height of the tropopause.~~(e) Plot of the ash mass corresponding to pixels shown in (d).~~

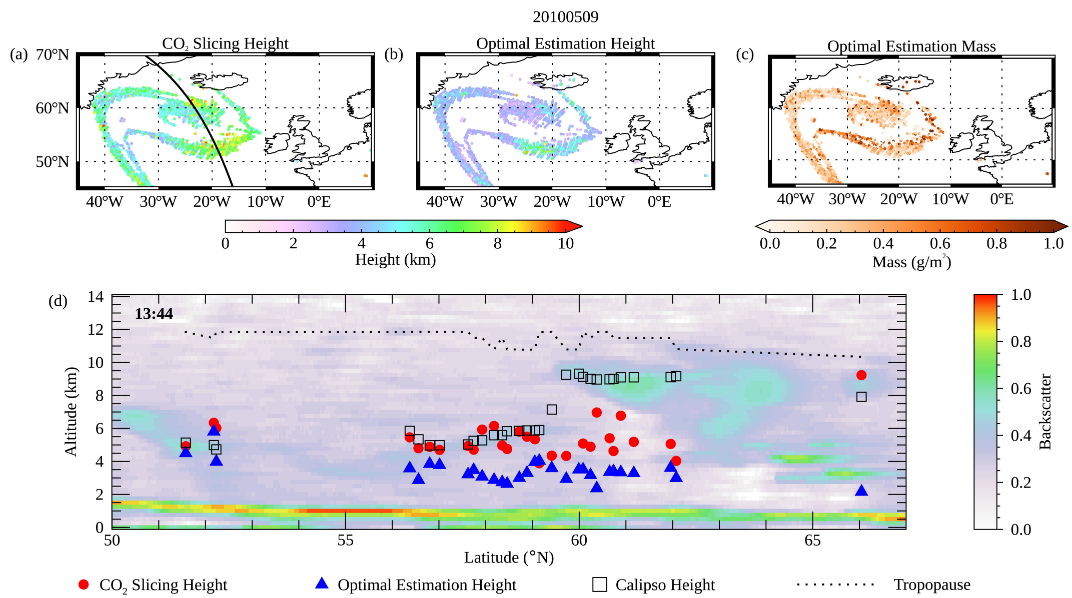

Figure 11. Same as figure 10 for 9th May 2010.

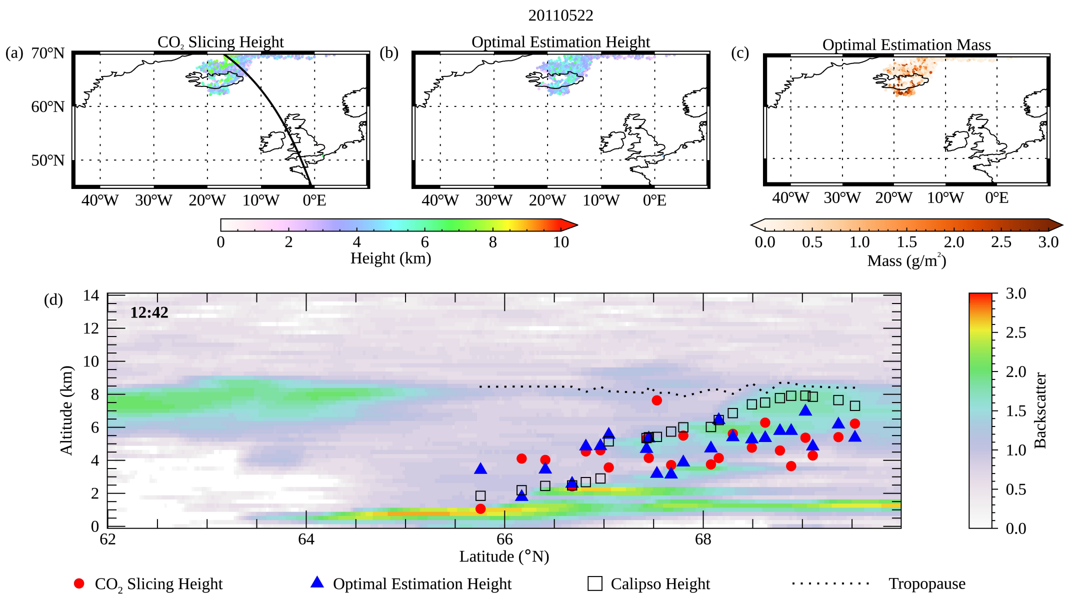

Figure 12. Same as figure 10 for 22^{nd} May 2011.

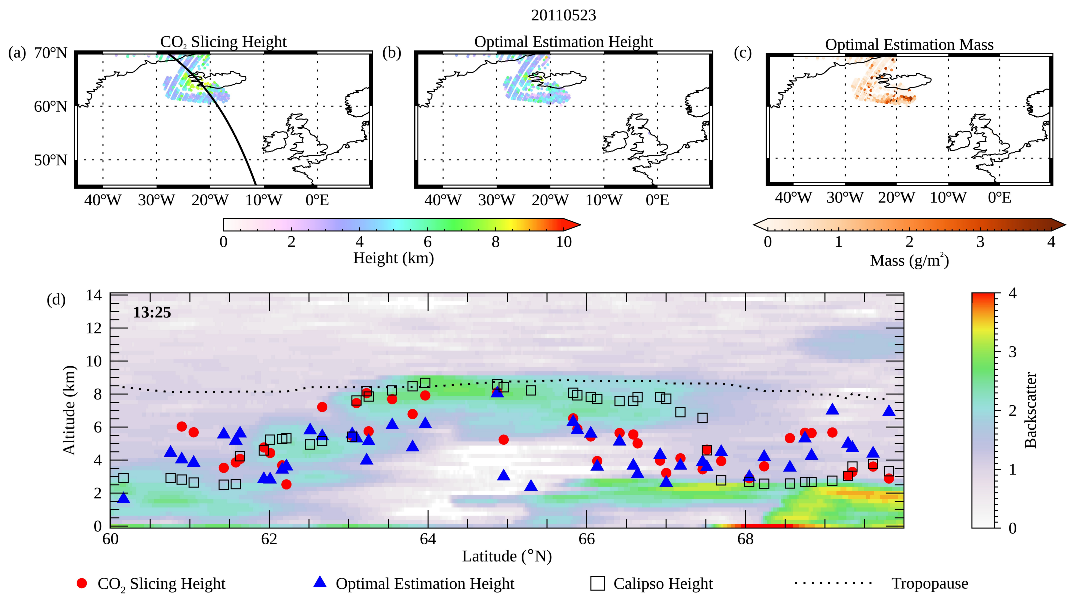

Figure 13. Same as figure 10 for 23rd May 2011.

Table 1. A summary of some of the existing methods for determining the height of volcanic ash clouds. Summaries can be found in Oppenheimer (1998); Prata and Grant (2001a, b); Zakšek et al. (2013)

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Method	Description	Examples in literature
Ground based methods		
Infrared camera	Infrared cameras measure the heat radiated off the ash cloud. This means the plume can be distinguished from its surroundings. The top of the plume can be identified and the height calculated by counting the number of pixels between the plume top and a reference point.	Patrick (2007); Sahetapy-Engel and Harris (2009); Webb et al. (2014); Bombrun et al. (2018)
Radar	A pulse of radio energy is emitted from a transmitter. This is reflected back off clouds (~~meteorological~~ aqueous or ash). This echo can be used to determine the cloud height.	Lacasse et al. (2004); Arason et al. (2011); Petersen et al. (2012)
Multiple platforms		
LiDAR	LiDAR is an active sensor which can be used on the ground as well as on aircraft or satellite platforms. The backscatter returned to the instrument can be used to infer the height of multiple cloud layers (including different types of cloud and ash). This is commonly used for validation of other methods.	Ansmann et al. (2010); Marenco et al. (2011); Winker et al. (2012); Vernier et al. (2013); Balis et al. (2016)
Satellite techniques		
Stereo view	This method requires two instruments viewing the cloud at the same time or a single instrument with two viewing angles (i.e. nadir and forward viewing). The resulting parallax can be used to determine the cloud height.	Prata and Turner (1997); Zakšek et al. (2013)
Cloud shadow	The shadow cast by clouds can be identified in visible satellite imagery. Combined with knowledge of the satellite viewing angle and the position of the sun, this can be used to find the height of the cloud layer. Alternatively multiple images including the cloud's shadow can be used.	Holasek et al. (1996); Prata and Grant (2001b)
Cloud top temperature	The cloud top temperature measured by an infrared instrument (usually at 11 μm) is compared against a temperature profile (e.g. radiosonde or weather model) to obtain the height.	Holasek et al. (1996)
Backward trajectory Modelling	Method uses the vertical wind directions and backwards trajectory modelling to get vertical distribution of ash. This can then be used to obtain the flux.	Eckhardt et al. (2008)[1],Stohl et al. (2009)[2],Kristiansen et al. (2010)[1],Stohl et al. (2011)[2], Pardini et al. (2017, 2018)[1]
Radiance fitting	Spectra are forward modelled given certain atmospheric parameters. These spectra are compared against those measured by the instrument and this is used to determine the altitude	Ventress et al. (2016); Zhu et al. (2017)

[1]Example using SO_2 not ash

[2]Example using hydrofluorocarbons and hydrochlorofluorocarbon

Table 2. A summary of some of the previous applications of the CO_2 slicing technique.

Instrument	**Platform type**	**Examples**
AIRS	Satellite	Pangaud et al. (2009)
GOSAT	Satellite	Someya et al. (2016)
IASI	Satellite	Arriaga (2007)
ITPR	Satellite	Smith and Platt (1978)
MODIS	Satellite	Menzel et al. (1992); Richards (2006)*; Tupper et al. (2007)*; Menzel et al. (2008)
MODIS MAS	Airborne	Frey et al. (1999)
S-HIRS	Airborne	Holz et al. (2006)
VAS	Satellite	Menzel et al. (1983); Wylie and Menzel (1989)

AIRS- Atmospheric Infrared Sounder

GOSAT- The Greenhouse Gases Observing Satellite

IASI- The Infrared Atmospheric Sounding Interferometer

ITPR- Infrared Temperature Profile Radiometer

MODIS- Moderate Resolution Imaging Spectroradiometer

MODIS MAS- Moderate Resolution Imaging Spectroradiometer Airborne simulator

VAS- Visible Infrared Spin-Scan Visible Radiometer Atmospheric Sounder

*Studies applied to ash

Table 3. The channel ranges selected for the final application of the CO_2 slicing technique. In total 57 channels are used. Following Arriaga (2007) 900.50 cm^{-1} is used as the window channel used to calculate the effective emissivity

StepCO_2 Channel Range (cm^{-1}) (inclusive)	CO_2 Channel Range (inclusive) Reference Channel (cm^{-1})	Peak Sensitivity Range (hPa)	Number of Channels
1 700 - 703.5	715	110.25 - 314.00	15
3 706 - 710.5	715	328.75 - 478.00	19
4 713 - 713.5	725	442.00 - 496.75	3
5 718.25 - 719.5	728	133.75 - 441.75	6
6 720.5 - 721.5	728	21.00 - 496.50	5
7 729.75 - 731.75	735	535.25 - 639.25	9

Table 4. Summary of the percentage of accepted retrievals and the RMSE describing the difference between the true (simulated) and retrieved values

	No quality control		With quality Control	
Atmosphere	**RMSE (m)**	**Success Percentage**	**RMSE (m)**	**Success Percentage**
RTTOV Standard	706	91	424	64
Mid-Latitude Day	635	100	282	84
Mid-Latitude Night	635	100	282	84
Tropical Day	1483	100	141	72
Polar Summer	1271	95	777	29
Polar Winter	565	100	1553	97
All	988	97.7	777	71.9

Table 5. Statistics describing the comparison of the CO_2 slicing and optimal estimation scheme against the heights obtained with CALIOP

	CO_2 slicing			**Optimal Estimation**		
Volcano	**Number of pixels**	**Correlation Coefficient**	**RSME (km)**	**Number of pixels**	**Correlation Coefficient**	**RSME (km)**
Eyjafjallajökull	53	0.2	2.2	67	-0.1	3.2
Grimsvötn	65	0.5	2.1	69	0.3	2.4
All	118	0.4	2.2	136	0.1	2.8

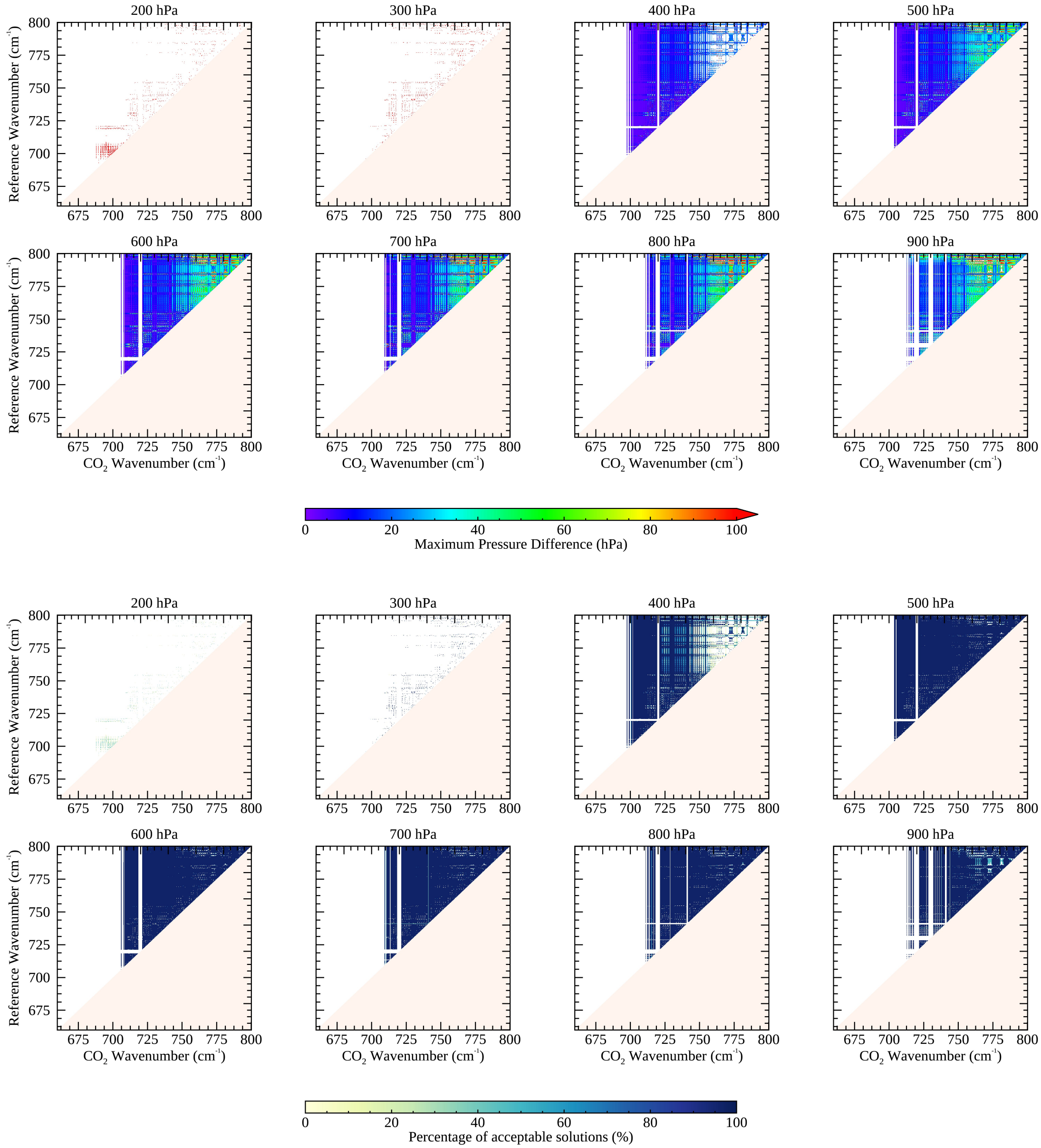

Figure A1. Simulation results for an RTTOV default atmosphere. The top two ~~line~~ rows shows the maximum difference between the true (simulated) and retrieved pressures grouped into the different pressure levels. Each level consists of ash optical depths ranging between 5 and 15 and effective radius between 5 and 10 μm. The bottom two ~~lines~~ rows show the percentage of accepted retrievals (i.e. the percentage of cases where there is an intersection between Eq. 1 and 2, and where all quality control criteria are met).

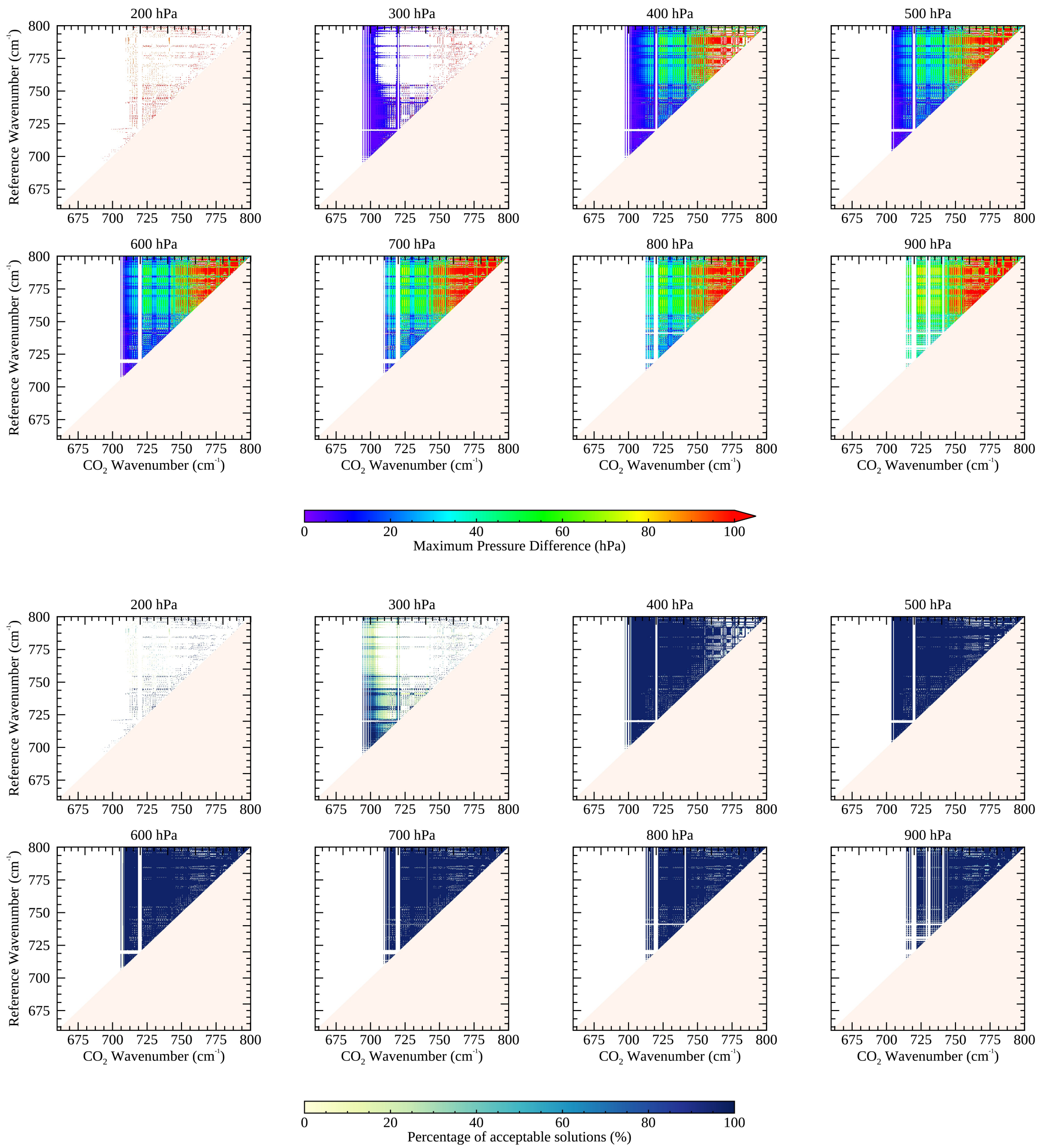

Figure A2. Same as figure A1 for a mid-latitude day atmosphere

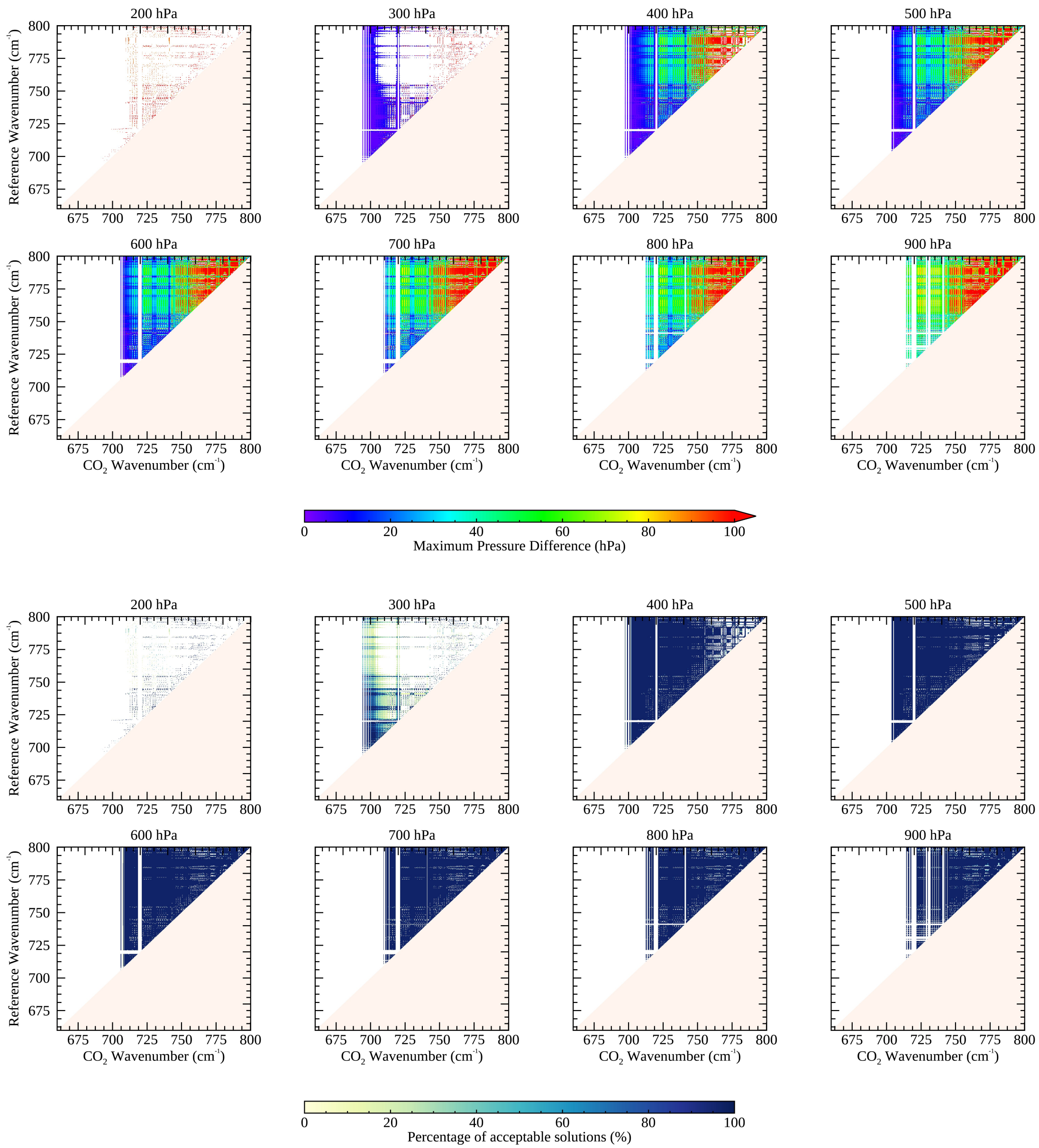

Figure A3. Same as figure A1 for a mid-latitude night atmosphere

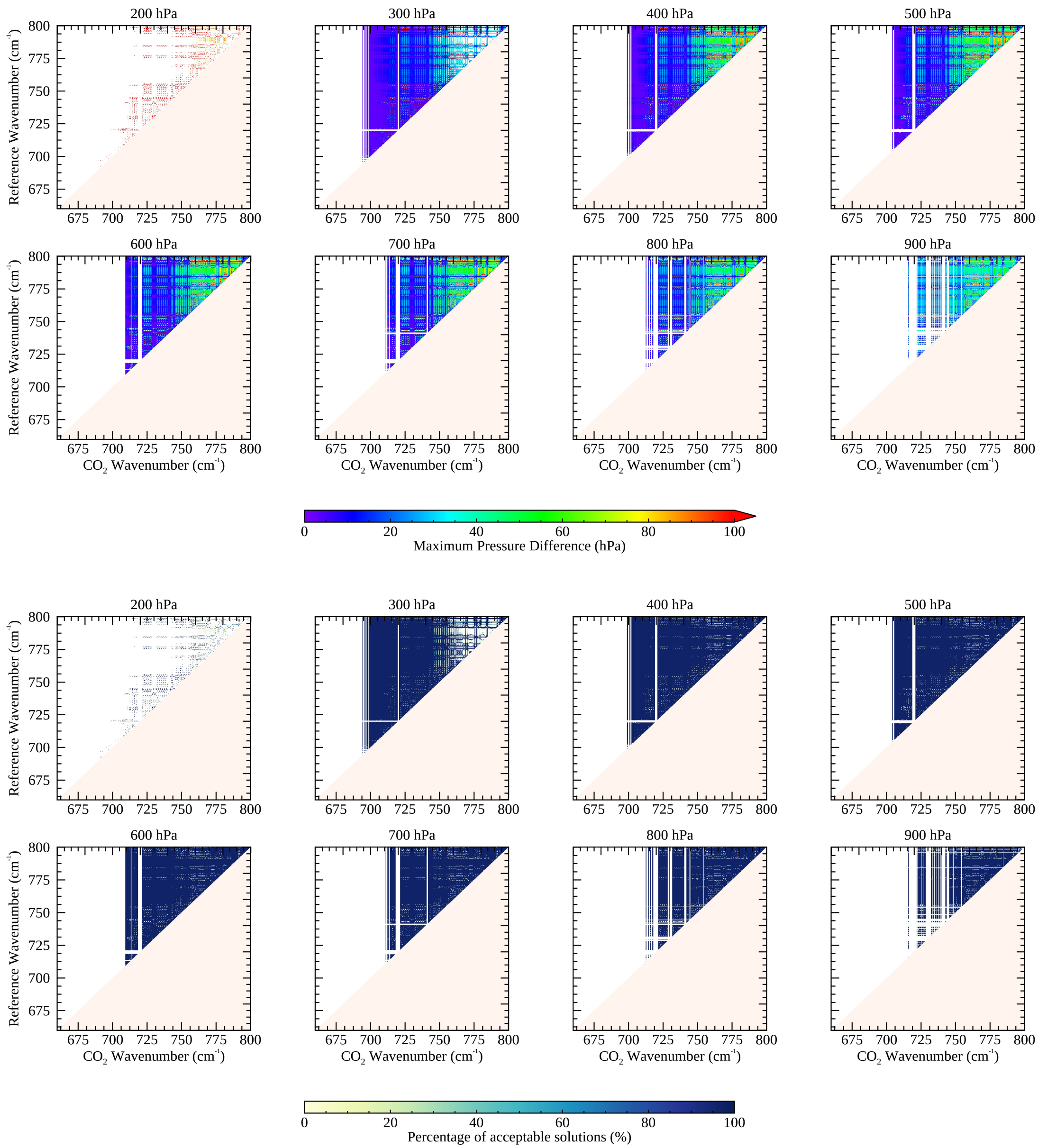

Figure A4. Same as figure A1 for a tropical atmosphere

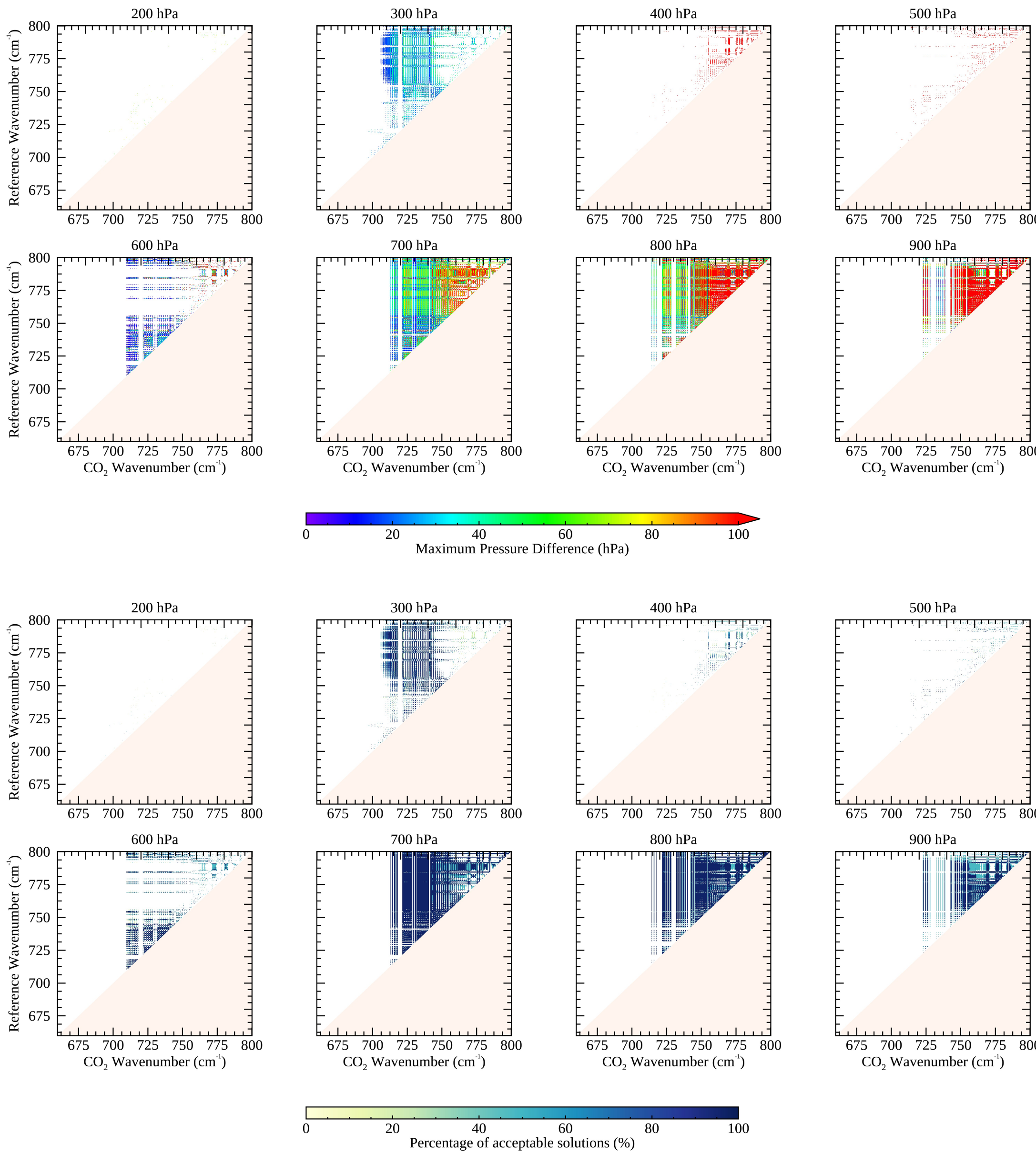

Figure A5. Same as figure A1 for a polar summer atmosphere

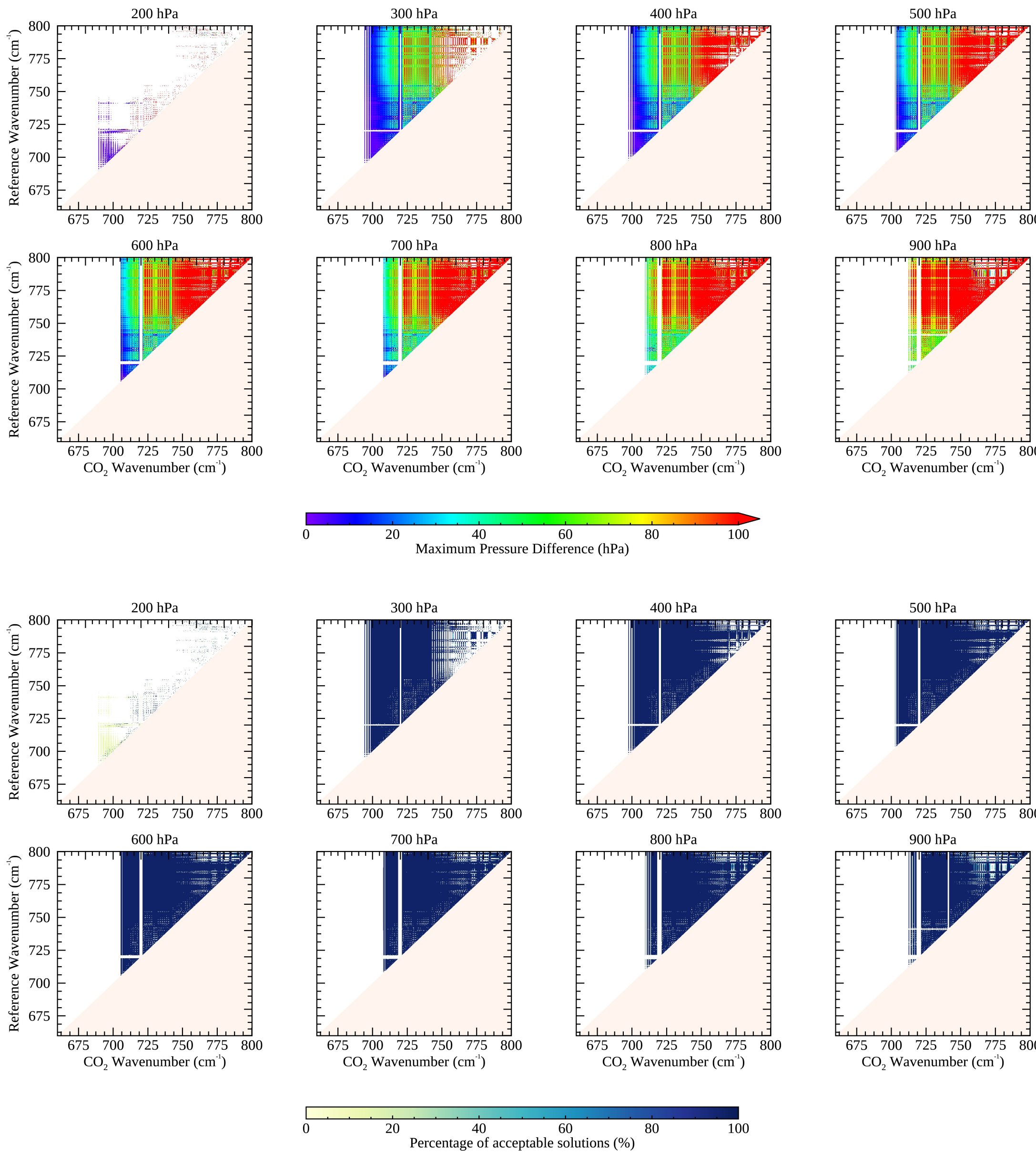

Figure A6. Same as figure A1 for a polar winter atmosphere

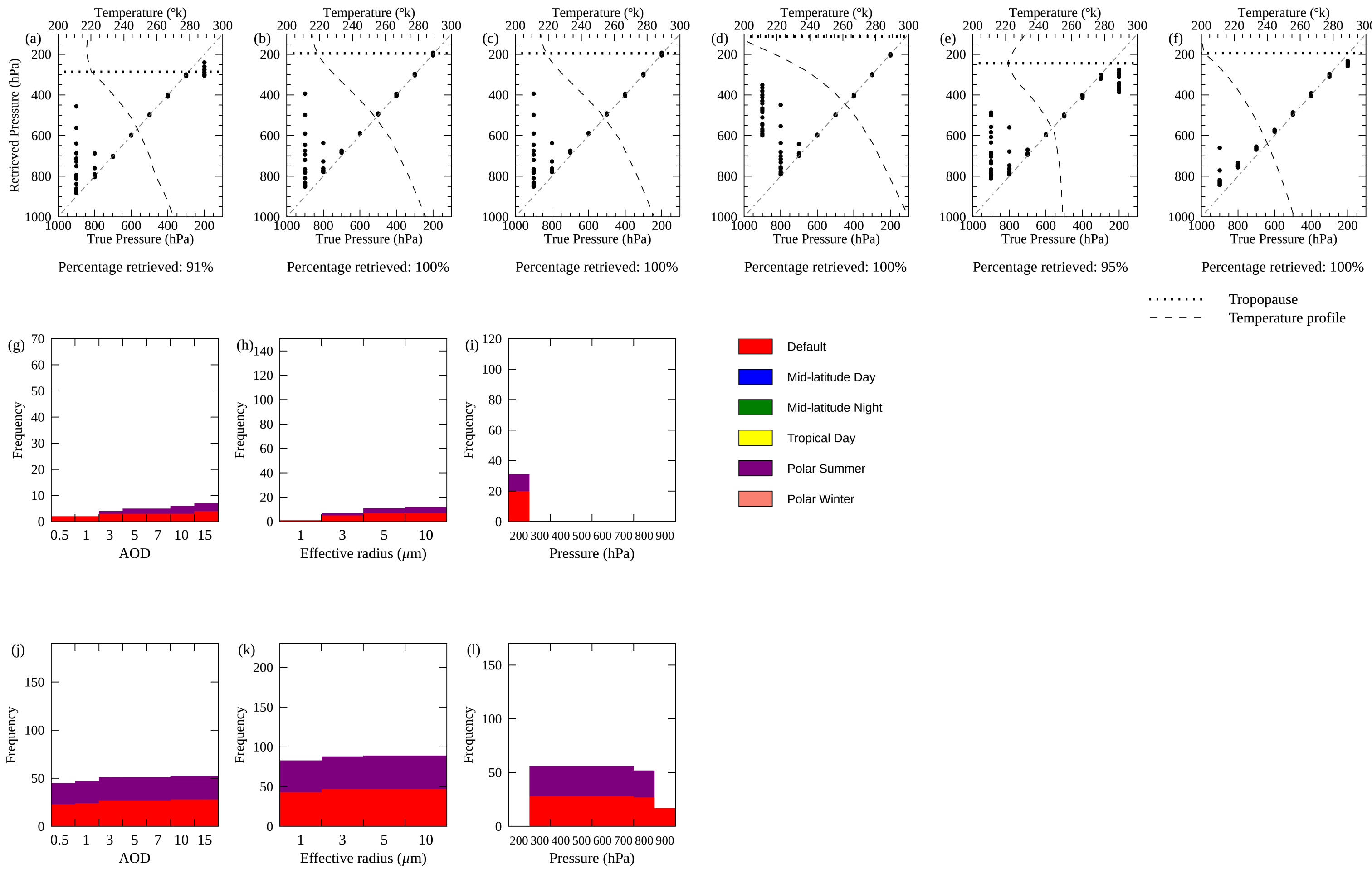

Figure A7. ~~Final CO_2 slicing pressure results for RTTOV simulated ash spectra (a total of 224 spectra per atmosphere). The plots show the true (simulated) pressure plotted against the CO_2 slicing retrieved value for the six different atmospheres. (~~Same as figure 6 without a ~~) RTTOV default atmosphere (high latitude), (b) Mid-latitude day, (c) Mid-latitude night, (d) Tropical day, (e) Polar summer (f) Polar winter. No~~ quality control ~~has been~~ applied.~~In this case, the simulated spectra include the following ash properties: ash optical depth ranging between 0.5 and 15, ash effective radius ranging between 1 and 10 μm and pressure values between 200 and 900 mb. Below each plot is a value indicating the percentage of successful retrievals. (g) The frequency of ash optical depths for which the CO_2 slicing technique was unable to return a height value. (h) Same as (g) for the effective radius. (i) Same as (g) for the ash cloud pressure.~~