Dear Editor,

we submitted the revised version of our manuscript:
‚Investigation of 3D radiative transfer effects for UV / vis satellite and ground based observations of volcanic plumes'

We followed almost all of the suggestions of the reviewers as described in detail below and in the comments in the pdf file attached to our replies to reviewer Christoph Kern:
(https://editor.copernicus.org/index.php?_mdl=msover_md&_jrl=400&_lcm=oc108lcm109w&_acm=get_comm_sup_file&_ms=106327&c=238151&salt=391523280658309828)
We also give clear reasons in the cases where we did not follow the reviewer's suggestions.
The changes are also marked in the manuscript file with tracked changes.

Below our responses to the reviewer's comments are marked in blue
The modified text of the revised manuscript is marked in red.

Best regards,

Thomas Wagner

Reviewer #3

General comments

The paper investigates the impact of 3D effects on satellite observations (SO_2, BrO and IO) of volcanic plumes by applying the 3D radiative transfer model TRACY-2 to highly idealized plumes. Four effects are investigated (horizontal light mixing, saturation effects, geometric effects and plume side effects) and it is found that especially the first two effects significantly bias SO_2 retrievals when considering only 1D radiative transfer. The paper provides substantial contribution to the scientific progress and is well within the scope of AMT.

The paper is quite unbalanced between the first part (Section 1 and 2) and the second part (Section 3-8). The authors introduce and discuss in detail scenarios for SO_2, BrO and IO for wavelengths of 313, 340 and 440 nm, respectively (Sect. 2.1.2-4). However, BrO and IO are not mentioned in the results, discussions and conclusions (besides figures are generally provided for these three wavelengths). Indeed, BrO and IO are mentioned last in line 315 at the beginning of the results (Section 3) and never mentioned afterwards. To add some confusion, the authors also discuss SO_2 retrievals at 313, 324, 332 and 370 nm in the context of the saturation effect, which adds a second wavelength dependency of the results. I think it is necessary to better balance the two parts. My suggestion would be to focus on SO_2 only in the paper for wavelengths 313 to 370 nm, remove BrO and IO from Sections 1 and 2, and simplify figures focusing mainly on the wavelength range relevant for SO_2 retrievals (e.g. 313, 340 and 370 nm). I would still find it important and interesting to discuss implications for BrO and IO (and other trace gases) in the conclusions. I think that these changes would results in a more concise and focused introduction and overall paper.

Author reply:
We thank this and another reviewer for the hint that the discussion and results are unbalanced between the first and second part.
Partly, this imbalance is a consequence of the fact that the saturation effect is only relevant for the SO_2 measurements. Thus a strong focus of the results and dicussion is on SO_2.
Also, part of this impression was probably caused by the fact that often only the wavelength (e.g. 440 nm for IO) was mentiond without explicit mentioning of the corresponding molecule.
Both reviewers recommend that the focus of the paper should therefore be reduced to SO_2.
However, we prefer to keep the BrO and IO results in the main part of the paper, and instead improve/complete the discussion of the BrO and IO results in the second part.
The main reason to keep the BrO and IO results in the main part is that our paper deals with the fundamental consequences of 3D RTM effects. And the most fundamental 3D effect is the light mixing effect, which strongly depends on wavelength. This wavelength-dependence is well covered by the three selected trace gases SO_2, BrO ad IO. Also the geometric effects and side scattering effects systematically depend on wavelength because of the different penetration depths of the incoming solar radiation on.
The reviewer also criticises that the two wavelength dependencies (1. for the three selected trace gases, and 2. for SO_2 at different wavelengths) leads to confusion.
Here it is important to point out that both sets of wavelengths were chosen to investigate two different effects (wavelength dependence of Rayleigh scattering and wavelength dependence of the SO_2 absorption cross section.
To make the choice of both sets of wavelengths more clear, the following text was added in section 2.1:
‚The scenarios for SO_2, BrO, and IO were chosen, because these trace gases were already observed in volcanic plumes. Moreover, the corresponding wavelengths cover the spectral range from about 310 to 440 nm, over which the probability of Rayleigh scattering strongly changes (by a factor of 4). Thus the light mixing effect (and also the geometric effect and the side scattering effect) are expected to differ substantially for the chosen wavelengths. A second set of wavelengths is selected for the SO_2 scenarios (4 wavelengths covering the spectral range from about 313 to 370 nm, for details see section 2.14). But here we are not primarily interested in the effect of the wavelength dependence of Rayleigh scattering, but rather on the strong wavelength dependence of the SO_2 absorption cross section. This second set of wavelengths is used to study the saturation effect for SO_2.'

We also added more discussion of the BrO and IO results in the revised version of the paper and we added the molecules to the corresponding wavelengtsh in most figures and many places in the text.

Specific comments

Reviewer comment:
L64: Suggest adding "TROPOMI ground pixel (e.g., 1 x 1 km²)"

Author reply:
Many thanks. The information was added

Reviewer comment:
L378ff: Please check percentage numbers as I get, for example, 7% instead of 8% (3.2/43.6) but these might be rounding errors.

Author reply:
Many thanks for this hint. You are right. 8% was replaced by 7%.

Reviewer comment:
L507f: It is still possible to have SZA = VZA and to pass the plume only once for the case of different azimuth angles.

Author reply:
Many thanks for this hint. We replaced ‚cases with SZA ≠ VZA' by ‚such cases'

Reviewer comment:
Figure 8: What do the error bars represent in the figure?

Author reply:
We added the following information to the figure caption (and also to the figure caption of the new Fig. A3.2:
‚The error bars represent the standard deviation calculated from 40 individual simulations.'

Technical corrections

Reviewer comment:
L299: "For such measurements, horizontal light paths can also play …"

Author reply: corrected

Reviewer comment:
L313: "Fig. 4" -> "Figure 4"

Author reply: corrected (now Figure 5)

Reviewer comment:
L383: Maybe change "considered here (see Table 3)" to "considered in Table 3", as the first version can imply that your conclusion for item c is found in Table 3, but your conclusion is actually supported by the following paragraph and Figure 10.

Author reply:
Many thanks for this hint. We changed the text as suggested.

Reviewer comment:
L419: Add "%" after 9.

Author reply: corrected

Reviewer comment:
L547: I guess it should be "50° in forward and backward direction" as in Figure 18.

Author reply:
corrected

Reviewer comment:
Figure 3: It is probably an imaging artefact, but I also see blue and purple dots in the figures.

Author reply:
Indeed there are also blue dots in the figure. They represent rotational Raman scattering events. We added this information to the figure caption.

Reviewer comment:
Figure 4: Number for VZA is missing.

Author reply:
Many thanks for this hint. The information (VZA: 0°) was added.

Reviewer comment:
Figure 17: The caption does not mention SO_2 and BrO, but only IO.

Author reply:
The figure caption was corrected accordingly.

Referee #2

The manuscript "Investigation of 3D-effects for UV/vis satellite and ground based observations of volcanic plumes" investigates the impact of the 3D structure of volcanic plumes on the satellite and ground based retrieval of trace gas species in the UV for which usually 1D scenarios are assumed. In detail, four effects are investigated that partly have a strong effect on the retrieved VCDs, especially for high spatial resolution instruments like e.g. TROPOMI. This paper is particularly interesting for the UV trace gas-retrieval community that observes narrow plumes of volcanic and anthrophonic sources and is well within the scope of AMT.

General comments

Although in the first part of the paper BrO and IO are introduced, the second part focuses mainly on the short wavelength UV range covering SO_2. I would therefore

suggest to only focus on SO2 in the paper and show some more results (see my comments below) and replace the 440nm results with the 370nm results
For some of the effects investigated you mainly focus on a plume around 5-6 km and sometime 10-11km, whereas other effects (i.e. the Plume side effect) only considers a plume on the ground. It would be nice to see (or discuss) the impact of each effect as a function of plume height. What I would like to see (if possible) a high-altitude plume around 15-17km in addition.

Author reply:
We thank this and another reviewer for the hint that the discussion and results are unbalanced between the first and second part.
Partly, this imbalance is a consequence of the fact that the saturation effect is only relevant for the SO_2 measurements. Thus a strong focus of the results and dicussion is on SO_2.
Also, part of this impression was probably caused by the fact that often only the wavelength (e.g. 440 nm for IO) was mentiond without explicit mentioning of the corresponding molecule.
Both reviewers recommend that the focus of the paper should therefore be reduced to SO_2.
However, we prefer to keep the BrO and IO results in the main part of the paper, and instead improve/complete the discussion of the BrO and IO results in the second part.
The main reason to keep the BrO and IO results in the main part is that our paper deals with the fundamental consequences of 3D RTM effects. And the most fundamental 3D effect is the light mixing effect, which strongly depends on wavelength. This wavelength-dependence is well covered by the three selected trace gases SO_2, BrO ad IO. Also the geometric effects and side scattering effects systematically depend on wavelength because of the different penetration depths of the incoming solar radiation on.
To make the choice of wavelengths more clear, the following text was added in section 2.1:
‚ The scenarios for SO_2, BrO, and IO were chosen, because these trace gases were already observed in volcanic plumes. Moreover, the corresponding wavelengths cover the spectral range from about 310 to 440 nm, over which the probability of Rayleigh scattering strongly changes (by a factor of 4). Thus the light mixing effect (and also the geometric effect and the side scattering effect) are expected to differ substantially for the chosen wavelengths. A second set of wavelengths is selected for the SO_2 scenarios (4 wavelengths covering the spectral range from about 313 to 370 nm, for details see section 2.14). But here we are not primarily interested in the effect of the wavelength dependence of Rayleigh scattering, but rather on the strong wavelength dependence of the SO_2 absorption cross section. This second set of wavelengths is used to study the saturation effect for SO_2.'

We also added more discussion of the BrO and IO results in the revised version of the paper and we added the molecules to the corresponding wavelengtsh in most figures and many places in the text.

As suggested by the reviewer we also added and discuss simulation results for other plume heights, especially also for a high plume between 15 and 16 km. Some of these results were added to existing figures (Fig. 5, Fig. 10, Fig. 12, Fig. 20). For other results new figures were added in the new appendix 3 (Fig. A3.1, Fig. A3.2, Fig. A3.3, Fig. A3.4, Fig. A3.5, Fig. A3.6).

Detailed comments

Reviewer comment:
Abstract, Line 25: „systematic underestimation" of which quantity? AMF? VCD? Please add

Author reply:
We changed the text to:
‚Especially the first two effects can lead to a strong and systematic underestimation of the retrieved trace gas content if 1D retrievals are applied...'

Reviewer comment:
Section 1, 83: "the true plume amount" – what amount do you mean, i.e. which quantity? Mass?

Author reply:
We changed the text to:
‚3D effects can cause an underestimation of the true trace gas content of the plume by more than....'

Reviewer comment:
Section 2, Line 164: "one grid cell from 555 km to 20°" – it is a bit confusing to suddenly switch from km to degrees – suggest to use km instead of 20°

Author reply:
Many thanks for this hint! We changed 20° to 2222 km.

Reviewer comment:
Section 2, Line 166: "The surface albedo was set to 5%" Please add a short justification why you use this albedo and which kind of surface this would represent.

Author reply:
We added the following information to the text:
‚This value was chosen, because typical albedo values in the considered wavelength ranges over volcanic areas are close to this value (see Fig. A1.1 in appendix A1). The exact choice of the surface albedo is, nevertheless, not critical, because the ratio of AMFs for narrow plumes and those for horizontally extended plumes hardly depends on the surface albedo (see Fig. A1.2 in appendix A1).'

The two new figures (Fig. A1.1 and Fig. A1.2) were added to the new appendix A1.

Reviewer comment:
Section 2, Line 171: "rectangular FOVs corresponding to the nominal ground pixel sizes of the different satellite instruments are used". Perhaps add in a table the

ground-pixel diameters of the different instruments such that the reader can compare between the instruments and the narrow FOV.

Author reply:
We added a hint in the text to table 1, where the ground pixel sizes of the different instruments are listed.

Reviewer comment:
Section 2, Line 224: Would it be possible to also add a high-altitude plume at 15-16km?

Author reply:
We performed additional simulations for plumes at 15 – 16 km as suggested by the reviewer. Now 4 heights are considered for the light mixing effect and the saturation effect, which are the most fundamental 3D effects. Part of these new results are added to existing figures (Fig. 5, Fig. 10, Fig. 12, Fig. 20). Others were added as new figures in the appendix 3 (Fig. A3.1, Fig. A3.2, Fig. A3.3, Fig. A3.4, Fig. A3.5, Fig. A3.6).

We added the following explanation to section 2.1.1:
‚For most simulations (light mixing effect and saturation effect) we investigate plumes in four altitude ranges: 0 - 1 km, 5 - 6 km, 10 - 11 km, and 15 - 16 km. For the study of the geometric effects and the side scattering effects only plumes at specific heights are chosen to illustrate the general dependencies'

Reviewer comment:
Section 2.1.2 & 2.1.3: Suggest to remove these and focus on SO2 in the following, see my general comment above

Author reply:
As explained in more detail above, we prefer to keep the simulations for BrO and IO in the main part of the text.

Reviewer comment:
Section 2.1.4 Line 270: The La Palma eruption occurred from September-December 2021, so I would either remove "Summer" or replace with "September to December 2021".

Author reply:
Many thanks for this hint! We replaced ‚Summer' by ‚September to December 2021'.

Reviewer comment:
Figure 6: Although the figure shows the SO2 fit ranges of the "MPIC analysis", this is nowhere described or mentioned in the text. Please remove this from the figure

Author reply:
We deleted the SO_2 fit ranges of the MPIC retrieval from the figure.
(note that Fig. 6 was shiftet to be the new Fig. 4)

Reviewer comment:

Figure 8: Can you also show the results for different plume heights in the plot and/or different wavelengths?

Author reply:
The results for all combinations of the 3 wavelengths and 4 plume heights were added as Fig. A3.2 to the new appendix A3. They are also discussed in section 3.1.

Reviewer comment:
Figure 10: Can you also show the results for different plume heights in the plot?

Author reply:
The results for the other 3 plume heights were added to Fig. 10.
In section 3.2 we also added the following discussion:
‚For plumes at higher altitudes, the underestimation becomes smaller because the probability of multiple scattering due to Rayleigh scattering decreases and thus the differences of the AMFs for narrow and horizontally extended plumes become smaller.'

Reviewer comment:
Figure 13 and Section 4.0 Line 429: You refer to scenarios "strong,1" and "strong,4" and also use this in the title of the plots- this is confusing since one automatically asks, what about strong,2 and strong,3. Suggest to remove this rather arbitrary scenario naming

Author reply:
We agree that the naming is a little bit confusing. Nevertheless, we think it is still meaningful to separate the SO_2 scenarios into weak and strong scenarios. The strong scenarios are then further subdivided into 4 scenarios to cover the range of realistic SO_2 amounts in volcanic plumes.
To minimise possible confusion, we added the following explanation to the figure caption of Fig. 13:
‚Here only the scenarios 'weak, 'strong,1' and 'strong,4' are considered to illustrate the transition from cases with no saturation to cases with medium or strong saturation. The additional intermediate scenarios 'strong,2' and 'strong,3' are later also used for the quantification of the saturation effect for the different plume extensions (Fig. 14) and satellite instruments (Fig. 15).'

Reviewer comment:
Section 4 Line 438-439: "Accordingly, with increasing plume height a stronger reduction of the observed radiance for plumes with high SO2 amounts is found" Where do I see this? Only one plume height result is shown.

Author reply:
Many thanks for this hint!
The effect is seen in the comparison of the results for different plume heights (new Fig. A3.4).
We added this information to section 4.

Reviewer comment:

Section 4.1 Line 456. Perhaps add a sentence here about the thresholds used to switch to other fit windows.

Author reply:
The following information is added to the text:
‚The threshold values are $4 \cdot 10^{17}$ molec/cm² for the switch from fit range 1 to 2 and $6.7 \cdot 10^{18}$ molec/cm² for the switch from fit range 2 to 3'

Reviewer comment:
Figures 18: Why is the peak observed in radiance not at the same distance as for the AMF? What is the VZA for the bottom AMF plots as a function of SZA?

Author reply:
We added the following explanation to the text:
‚Especially for IO at 440 nm the normalised radiances and AMFs for the narrow FOV show complex dependencies on the viewing angle. Moreover, while the enhanced values of the radiances and the AMFs are found at similar viewing angles, there are also systematic differences in the details of their viewing angle dependencies. These are caused by the different sensitivities of both quantities on the atmospheric light paths. While the radiance mainly depends on the probability of scattering (on molecules and aerosols), the AMF also strongly depends on the length of the light path through the trace gas plume. These dependencies can be e.g. seen in the upper left part of Fig. 18a, where the maximum of the radiance is found at about 1.5 km, while the maximum AMF is found at a distance of about 0.8 km from the plume center.'

The VZA for the bottom panels is 60°. This information was added to the figure caption and to the text.

Reviewer comment:
Section 6: You have investigated this effect only for a plume located at the surface – please also add results for other plume heights

Author reply:
Here we did not follow the suggestion of the reviewer, because our intention was to chose a scenario, where the side scattering effect can be studied without interference from other 3D effects, especially the geometric effects. To make this more clear, we added the follwing text at the beginning of chapter 6:
‚We chose this low plume altitude, because for such plumes the side scattering effect can be investigated without interference with the geometric effects (section 5). Of course, the side scattering effect also affects plumes at higher altitudes (in addition to geometric effects).'

Also the title was changed to
'Plume side effects for low plumes'

Reviewer comment:
Section 8: Line 625: "…to a strong and systematic underestimation if 1D…". Underestimation of which quantity? Please specify

Author reply:
To make this more clear we changed the sentence to:
‚Especially the first two effects can lead to a strong and systematic underestimation of the true trace gas content of the plume if 1D assumptions are used in the data analysis (up to more than 50% for the light mixing effect, and up to 100% for the saturation effect).'

Reviewer comment:
Section 8, Line 627: Perhaps specify for which conditions a 100% saturation or 50% light mixing effect occurs.

Author reply:
We added the following text to section 8:
‚In such cases, wavelength-dependent correction factors (according to the results in Fig. 10) have to be applied to the results if a 1D AMF is used. The strongest underestimation (>50%) caused by the light mixing effect occurs for observations at short wavelengths and plumes below 10 km.
The saturation effect can lead to a further strong underestimation of the analysis results for cases with strong SO_2 absorptions. The strongest saturation effect occurs for observations at short wavelengths and for narrow plumes with high SO_2 VCDs (e.g. about > 95% underestimation for observations at 313 nm of a 1 x 1 x 1 km³ plume with SO_2 VCDs > $2.5 \cdot 10^{19}$ molec/cm²).

Christoph Kern (Referee)

In this manuscript, the authors investigate how 3D radiative transfer in and around volcanic plumes impacts remote sensing observations from space and, to a more limited capacity, from the ground. They find that light mixing plays a fundamental role in measurements made at spatial resolutions corresponding to the horizontal extent of volcanic plumes or smaller. In this situation, light that has not passed through (or has only partially passed through) the volcanic plume is mixed with light coming through the plume, thus reducing the total absorption signature stemming from trace gases in the plume. The study focuses on the UV/vis wavelength region where SO2, BrO, OClO, IO, and NO2 are typically measured, and thus has relevance to anthropogenic pollution plumes as well as volcanic plumes.
Besides light mixing, the study also investigates the effect of strong SO2 absorption on satellite remote sensing measurements and geometric effects related to non-vertical illumination/observation angles. Depending on the specific plume SO2 loading and the geometry of the measurement, these also play an important role. Finally, the last section of the manuscript deals with ground-based measurements, and in particular their sensitivity to the horizontal extent of overhead plumes. Here, the authors find that groundbased measurements of trace gas VCDs may be overestimated for very large gas plumes (converging towards elevated trace gas layers) unless realistic radiative transfer is considered.

The effects examined in this study are of fundamental importance to the remote sensing community. Although some previous articles have presented favorable comparisons between ground-based and satellite-based remote sensing observations of volcanic gas plumes, these have often relied on very limited and carefully selected datasets. In fact, the comparison between ground-based and space-based measurements is seldom perfect, and often the two measurement geometries yield quite different results. This article points out some important physical effects, many of which interestingly don't become relevant until the spatial resolution of the satellite instrument is improved to the point where it is on the same order of magnitude as the spatial extent of volcanic plumes, a circumstance that was realized with the successful launch of the TROPOMI instrument. The article is wellwritten, logically organized, and relatively easy to follow. I recommend it be published in Atmospheric Measurement Techniques once the mostly minor comments listed in the attached annotated manuscript are addressed, and the authors consider the following:

My only significant comment relates to section 7 on ground-based measurements. After spending 14 pages discussing a large variety of effects inherent in space-based remote sensing observations, the authors write less than one page about ground-based measurements. Unsurprisingly, this section is of limited value as it stands. A few issues to consider are listed below:

Author reply:
We thank Christoph Kern for his detailed, constructive and positive assessment of our study.
It is true that the chapter on gound based measurements is rather short. In fact it discusses only the light mixing effect, because the light mixing effect is most fundamental 3D effect. The reviewer suggested two ways to deal with that situation (see below), from which we chose the second one: ‚If, instead, the authors feel strongly about keeping the ground-based modeling results in the main paper, please ensure that the limited validity of the results is clearly stated (zenith-facing measurements, low SZAs, aerosol-free plumes, weak SO2 absorption) and that the study is only meant to make the point that ground-based observations are also sensitive to horizontal plume extent.'

We now make clear in chapter 7 that:
a) the ground based results address only the light mixing effect, because it is the most fundamental 3D radiative transfer effect.
b) the results cover only a limited set of scenarios (zenith viewing angle, low SZAs, aerosol-free plumes, weak SO_2 absorption).
c) the results can not be directly compared to those in Kern et al. (2010), because Kern et al. (2010) assumed a plume with infinite extension in one horizontal direction, while we assume confined plumes in all dimensions.

Section 7 was thus almost completely re-written. In the revised version it contains the following text:
‚3D effects are not only important for satellite observations of volcanic plumes, but also for ground based observations. In this section we briefly deal with this topic but only discuss the light mixing effect for ground based observations, because it is the most fundamental 3D radiative transfer effect. Kern et al. (2010) investigated 3D effects for ground based observations of volcanic plumes. Their focus was on the

effect of direct sun light scattered into the line of sight of the instrument without having crossed the plume before, which is referred to as light dilution effect (see also Millán, 1980). The light dilution effect is usually the dominant effect for such observations. While the exchange of light between the plume and outside the plume by multiple scattered photon was also included in their simulations, its contribution to the 3D radiative effects was not explicitly discussed.

In our simulations for ground based observations we therefore consider two scenarios (see Fig. 20):

a) ground based observations for zenith direction and very small SZA (here 0.3°): In this scenario the single-scattered light must have crossed the narrow plume before it is scattered into the instrument (like for the satellite observations with SZA = VZA = 0, see section 3). Thus no direct sun light is scattered into the line of sight of the instrument without having crossed the plume before. These results can be directly compared to the results for satellite observations.

b) ground based observations for zenith direction and slightly larger SZA (here 10°): In this scenario the single-scattered light has not crossed the narrow plume before it is scattered into the instrument (like for scenarios in Kern et al., 2010).

Note that for both scenarios single scattered light can also be scattered above the plume into the line of sight of the instrument, but these light paths do not fall under the definition of the light dilution effect. The results for both scenarios are shown in Fig. 21. For the first scenario, like for the satellite observations (Fig. 5), the strongest dependence is found for 313 nm, because the probability of Rayleigh scattering is largest for the short wavelengths. But the altitude dependence is opposite to that for the satellite observations. Here it is interesting to note that for the AMF ratio between narrow and horizontally extended plumes, no monotonous altitude dependence is found. This is caused by the non-monotonous altitude dependence of the 1D AMFs for horizontally extended plumes. Interestingly, the highest 1D AMFs are found for plumes at medium altitudes (here altitudes of 5 and 10 km). For plumes at lower altitudes, the effect of multiply scattered photons is reduced because of the low surface albedo. For higher altitudes, it is reduced because of the low probability of Rayleigh scattering with decreasing air density.

The results for the second scenario show even lower AMFs for the narrow plumes than those for the first scenario. This is caused by the direct sun light scattered into the line of sight without having crossed the plume before (similar as for the scenarios in Kern et al., 2010). However, a direct comparison of the results of Kern et al. (2010) to the results of this study is complicated, because of two reasons: first, Kern et al. (2010) assumed a plume with infinite extension in one horizontal direction, while we assumed confined plumes in all dimensions. Second, they compared the AMFs of their 3D radiative transfer simulations with the assumption of a geometric light path through the plume, while in our study we compare the 3D AMFs with the corresponding 1D AMFs for horizontally extended plumes. Despite these differences, the general dependencies of the results of Kern et al. (2010) and our results are very similar: the AMF can be strongly reduced by the 3D effects. While for plumes in close proximity to the ground, the AMF is similar to the geometric AMF (but smaller than the corresponding 1D-AMF), it strongly decreases with increasing plume altitude (or horizontal distance between instrument and plume). These results indicate that usually for ground based observations not only the dilution effect (as described in Kern et al. (2010) but also the light mixing effect is important.

The results of this study also indicate that the correction factors presented by Kern et al. (2010) are only valid for the chosen plume size (vertical extent of 500 m, horizontal extent in one dimension: 600 m, and infinite in the other dimension). An assumed change of the plume extent from 200 m to 4 km changes the AMFs by about 5 % (for 440 nm) and 30 % (for 313 nm). The dependence of the AMFs on the horizontal plume extent is probably smaller for more realistic plumes (like those in Kern et al., 2010). Nevertheless, for future analyses of ground based observations, the plume size should be also taken into account. It should be noted that our simulations cover only a limited set of scenarios (zenith view, small SZA, no aerosols, no strong SO_2 absorptions). While our results are useful to illustrate the general influence of the light mixing effect and dilution effect on ground based observations, they are not representative for all measurement situations. Simulations for a more comprehensive set of scenarios (including more realistic plumes) and covering also the other 3D effects should be performed in future studies.'

Reviewer comment:
It is difficult to understand the model scenarios without a sketch. E.g., if the SZA is set to 0 and the VZA is also 0, then the instrument would appear to be aimed directly at the sun – a seldom used (but interesting) geometry for volcanic plume measurements.
But the modeling results imply these were likely not direct-light observations. Please clarify this.

Author reply:
Many thanks for this hint! We added a sketch to the chapter (new Fig. 21) and clarified the role of direct sun light (see point above). It is correct that in our simulations only scattered light is considered (even if the viewing geometry allows direct light to be detected by the instrument). To avoid confusion, we now changed the viewing geometry from SZA of 0° to SZA of 0.3°. With that modification, the results are almost identical with the previous results for SZA=0°. However, for an SZA of 0.3° no direct sun light can reach the narrow field of view of the instrument.

Reviewer comment:
The simulations are compared to those run by Kern et al. (2010), but here it should be mentioned that the Kern et al. (2010) scenarios considered a plume of effectively infinite extent in one horizontal direction, which is different than the horizontally limited plumes studied here.

Author reply:
Many thanks for this hint! Indeed we overlooked this important detail. We clarified this fact in the revised version of the manuscript (see our answer above).

Reviewer comment:
The difference between "light dilution" and "light mixing" is highlighted, but it's not completely clear to me how these effects differ. In Kern et al. (2010), light dilution is defined as follows: "Besides the photons scattered behind the plume, some photons will also be scattered in the direction of the instrument between the instrument and the plume. These photons have not passed through the plume and therefore this contribution does not contain spectral absorption structures originating from plume constituents." Note, in particular, that there is no mention of the fact that the 'diluting' light must have been scattered only once on its way to the instrument. Light dilution can therefore also include light that has been scattered multiple times in the

atmosphere on its way to the instrument, so long as it has not passed through the plume.
In line 104, the authors of this study define light mixing as "part of the detected photons originate from air masses outside the observed ground pixel (and also from outside the trace gas plume)." This seems quite similar, though more tailored to a nadirviewing geometry. With this definition, I'm not quite sure how light mixing applies to the ground-based measurements, or how it compares to light dilution. If the authors want to refine the scope of the definition to include contributions from any light paths that are not equivalent to the instrument line of sight (whether they are in the plume or not), then I agree with their assessment that light dilution is a subset of light mixing, and that light mixing is perhaps a more general term. As it stands, I'm a bit confused by this comparison of light mixing and light dilution.

Author reply:
Many thanks for pointing these inconsistencies out!
The reviewer is right that with our original definition of the light mixing effect given in the introduction, the light mixing effect also covers the light dillution effect.
To clarify the defintion, we added the following discussion to section 7:
‚3D effects are not only important for satellite observations of volcanic plumes, but also for ground based observations. In this section we briefly deal with this topic but only discuss the light mixing effect for ground based observations, because it is the most fundamental 3D radiative transfer effect. Kern et al. (2010) investigated 3D effects for ground based observations of volcanic plumes. Their focus was on the effect of direct sun light scattered into the line of sight of the instrument without having crossed the plume before, which is referred to as light dilution effect (see also Millán, 1980). The light dilution effect is usually the dominant effect for such observations. While the exchange of light between the plume and outside the plume by multiple scattered photon was also included in their simulations, its contribution to the 3D radiative effects was not explicitely discussed.'

Reviewer comment:
While I agree with the assessment that the plume extent (both horizontal and vertical) plays a role in the UV/vis radiative transfer (and therefore the sensitivity) of groundbased remote sensing measurements, and that this fact is perhaps not adequately presented in the existing literature, I'm a bit wary of the results presented in section 7.
The authors write that "an assumed change of the plume extent from 200 m to 4 km changes the AMFs by […] 30% (for 313 nm)." This is true for the one scenario considered here – a square plume (more of a cloud really) that extends equally in both horizontal directions, a situation rarely encountered in actual ground-based measurements of volcanic plumes. The sensitivity of the AMF to plume altitude is not discussed, even though this is quite interesting as the results appear to indicate that there is an 'ideal' altitude at which ground-based instruments become most sensitive to large overhead plumes. (This appears to stem from the balance of scattering occurring both above and below the elevated plume?). Also, only SZAs of 0 and 10 degrees are investigated, although the AMFs for large, high plumes are surely sensitive to this parameter, and the majority of ground-based observations are made at higher SZAs.
And I guess that SO2 absorption was considered as weak?

Author reply:
We agree that the results only cover a small subset of possible scenarios and that the assumed plumes do not perfectly represent real plumes.
To make this more clear, we added the following text to the revised versions of our manuscript:
‚It should be noted that our simulations cover only a limited set of scenarios (zenith view, small SZA, no aerosols, no strong SO_2 absorptions). While our results are useful to illustrate the general influence of the light mixing effect and dilution effect on ground based observations, they are not representative for all measurement situations. Simulations for a more comprehensive set of scenarios (including more realistic plumes) and covering also the other 3D effects should be performed in future studies.'
We now also discuss the height dependence and added the following text:
‚…like for the satellite observations (Fig. 5), the strongest dependence is found for 313 nm, because the probability of Rayleigh scattering is largest for the short wavelengths. But the altitude dependence is opposite to that for the satellite observations. Here it is interesting to note that for the AMF ratio between narrow and horizontally extended plumes, no monotonous altitude dependence is found. This is caused by the non-monotonous altitude dependence of the 1D AMFs for horizontally extended plumes. Interestingly, the highest 1D AMFs are found for plumes at medium altitudes (here altitudes of 5 and 10 km). For plumes at lower altitudes, the effect of multiply scattered photons is reduced because of the low surface albedo. For higher altitudes, it is reduced because of the low probability of Rayleigh scattering with decreasing air density.'

Reviewer comment:
Finally, the role of aerosols and how they affect ground-based observations of the plumes presented here is not touched upon, even though aerosols were shown to have considerable influence on the satellite measurements, and it is known (e.g., from Kern et al. 2010) that they are major sources of uncertainty in ground-based measurements as well.

Author reply:
We agree and added the following text:
‚It should be noted that our simulations cover only a limited set of scenarios (zenith view, small SZA, no aerosols, no strong SO_2 absorptions). While our results are useful to illustrate the general influence of the light mixing effect and dilution effect on ground based observations, they are not representative for all measurement situations. Simulations for a more comprehensive set of scenarios (including more realistic plumes) and covering also the other 3D effects should be performed in future studies.'

Reviewer comment:
Taking these issues into account, I wonder what the best solution is. If ground-based measurements should be fully considered, the range of ground-based model scenarios would need to be expanded to include sensitivity studies of SZA, VZA, AOD, SSA, SO2 concentration, etc., similar in detail to what is given for the satellite measurements.
However, that would of course greatly expand the scope and length of the article.

Another option (this would be my recommendation) might be to keep the focus of the article on the space-based measurements, potentially remove ‘ground based’ from the title, move the ground-based sensitivity study to an appendix, and reference it farther up in the article where the influence of plume size on satellite-based measurements is discussed, e.g., writing something along the lines of “Ground-based remote sensing measurements were also found to be sensitive to horizontal plume extent, see Appendix B.” Then, a follow-up study could more fully investigate the 3D radiative transfer effects of ground-based measurements.
If, instead, the authors feel strongly about keeping the ground-based modeling results in the main paper, please ensure that the limited validity of the results is clearly stated (zenith-facing measurements, low SZAs, aerosol-free plumes, weak SO2 absorption) and that the study is only meant to make the point that ground-based observations are also sensitive to horizontal plume extent. (I think that is the main point, right?)

Author reply:
Many thanks for these suggestions! We followed suggestion #2 (see our responses above and the new section 7)

Reviewer comment:
The above (hopefully constructive) criticism of the ground-based measurement section should not take away from the fact that this article contains a wealth of extremely useful information. Understanding the processes described here represents the first step in consolidating space-based and ground-based measurements of volcanic gas plumes in a much more robust and comprehensive manner than was possible before. I appreciate the opportunity to comment on this important work.

Please also note the supplement to this comment:
https://amt.copernicus.org/preprints/amt-2022-253/amt-2022-253-RC3-supplement.pdf

Author reply:
Many thanks for these valuable additional comments! We followed almost all of them and explained our reasons in the cases where did not follow the suggestions. Our responses are added directly after the comments from the reviewer.