Atmospheric Measurement Techniques Discussions

Atmos. Meas. Tech. Discuss., referee comment RC3
https://doi.org/10.5194/amt-2022-253-RC3, 2022

Comment on amt-2022-253

Christoph Kern (Referee)

Referee comment on "Investigation of three-dimensional radiative transfer effects for UV–Vis satellite and ground-based observations of volcanic plumes" by Thomas Wagner et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2022-253-RC3, 2022

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 SO_2, 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 SO_2 absorption on satellite remote sensing measurements and geometric effects related to non-vertical illumination/observation angles. Depending on the specific plume SO_2 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 ground-based 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:

- 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.
- 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.
- 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 nadir-viewing 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.
- 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 ground-based 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 SO_2 absorption was considered as weak?

- 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.

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, SO_2 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 SO_2 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?)

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