Review of Griessbach et al., Infrared limb emission measurements of aerosol in the upper troposphere and lower stratosphere

Reviewer: Mike Fromm

Note: In this review I will refer to the author team together as "auth."

Review Synopsis

This manuscript addresses a very important topic in the study of the upper troposphere and lower stratosphere (UTLS), the satellite remote sensing of aerosol and the challenge/promise of aerosol type attribution. Auth make the case clearly that their objective is to treat both sides of the tropopause with equal fidelity. The UTLS is important for climate sensitivity (e.g. Solomon et al. (2011)) yet it is still a frontier of sorts because it represents a local minimum in accurate global measurements and because it is routinely occupied by water-ice clouds, which are confounding in terms of characterization and cloud/aerosol typing. Moreover, the true aerosol/cloud composition of the UTLS involves a rich mix of processes responsible for determining the gaseous and particulate profile, from (for example) in situ particle formation to eruptive/impulsive events like cumulonimbus convection and volcanic activity. Hence studies such as this are critically important and perfectly suited for AMT.

Auth are building on a foundation of MIPAS-related cloud and aerosol typing, and working their way down from the "free" stratosphere into the more complicated UTLS. The methods and results they report on here appear to show advances in MIPAS aerosol detection and classification. The approach, combining a theoretical and observation framework, is appropriate and good. In particular, the finding that they can discern vertically resolved water-ice, volcanic ash, and volcanic sulfate in the UTLS merits publication. However, because the global UTLS particulate composition is vastly more complex than this limited array, this work leaves important questions unaddressed and unresolved. Two examples illustrate the point. Mineral dust is known to be in residence at upper tropospheric altitudes (e.g. Husar (2001)). Forest fire smoke is episodically injected into the UTLS (Fromm et al., 2010). It is true that MIPAS has sampled several such events. For this paper to merit publication in AMT, a state-of-the-science accounting of the various UTLS particle types must be acknowledged and the impact of these particle types on MIPAS remote sensing must be addressed. In its current form, this manuscript falls short of this standard. Consequently, I recommend substantial additions before it can be accepted for publication in AMT.

So far, non-ice aerosol detection from infrared limb emission measurements is restricted to the stratosphere simply by assuming that particles in the stratosphere are aerosol and not ice (except for ice PSCs) or by assuming relatively high CI values (Sembhi et al., 2012). However, for stratospheric cirrus (Spang et al.,2015; Keckhut et al., 2005) or field-of-view effects that can also lead to high CI values these assumptions are not always valid.

What we have achieved in this paper is the first important step, namely the discrimination of ice clouds from other aerosol. Since the most likely and best defined aerosol candidates in the MIPAS data are volcanic aerosols (sulfate and ash), we focus on these volcanic aerosols and think that this is sufficient to demonstrate that the presented method is capable of distinguishing between ice clouds and non-ice aerosol.

We are very well aware that UTLS aerosol is much more than just volcanic aerosol. However, developing methods to unambiguously classify all possible UTLS aerosols is far beyond the scope of this paper. This paper is meant to do the first step of filtering out ice clouds and providing information on the location of aerosol, irrespective of the aerosol type. By applying this aerosol detection method to the whole MIPAS time series, we know that it captures sulfate aerosol, volcanic ash, mineral dust, wild fires, and non-ice PSCs. However, for developing methods to classify the aerosol types, separate studies focusing on one particular type would be required (e.g. for PSCs: Spang et al. 2003, 2004, 2005, 2012, Hoepfner et al. 2004, 2006, 2009; for volcanic ash: Griessbach et al. 2014). Methods and studies for mineral dust and wild fires are not available.

Hence,we consider further and better characterization of aerosol types as future work. In the revisied version this will be clarified.

I found myself unable to understand large portions of sections 3.1 and 3.2, on the method for aerosol detection and classification. My difficulty had to do with the sections' logic, clarity, and internal consistency. Since this represents the core of this AMT candidate, I suggest substantial rework to these parts before the paper can be accepted for publication. Details of my concerns will follow.

Next I will list the major, then minor concerns with the manuscript.

Major:

In the Introduction auth review the omnipresence of aerosol in the climate system. But they leave out some potentially important aspects of UTLS aerosols such as mineral dust presence in the uppermost troposphere (e.g. Husar et al., 2001; Cottle etal., 2013; Liu et al., 2013), upper tropospheric non-volcanic sulfates (Clarisse et al., 2012), and UTLS smoke (e.g. Fromm et al.,2010). To the extent that these and other particle types are likely present in the MIPAS record, it is essential to account for all UTLS pathways in the motivation for this paper.

We agree and will include a discussion in the introduction.

UTLS dust, smoke, and aerosols other than volcanic ash and sulfates are not included in the aerosol classification part of this paper. Given that the UT aerosol classification is a strong focus of this paper, it could be argued that dust, smoke, and anthropogenic pollution events dominate volcanic perturbations. Either they should be part of the scope of this work or auth need to expressly acknowledge/justify their exclusion.

Since this paper is intended to present an ice cloud filter (classification between aerosol in general and ice clouds) and does not aim at classifying all the various aerosol types it is beyond the scope of this work to include more aerosol types. However, we think it is more convincing to have at least two example aerosols (volcanic aerosol in our case) to show that there are indeed aerosols that exceed the thresholds and can clearly be discriminated from ice clouds.

Section 3.1: I describe below the details of my overall major concern with this section. Some of the items within this description can be considered minor but they are all together to make my point.

I don't understand how selecting a second window band, to distinguish AI from CI, is a strategic consideration. CI already has a window band in the denominator. Auth do not give a compelling reason on P4385 for substituting another window band for the 833/cm band in CI. They do state that they are aiming for altitude and seasonal independence, but they do not make the case that the CI's dependence on altitude and season are driven by the 833/cm window radiances. The apparent justification for choosing 960/cm is given in a new paragraph on P4386. If this is indeed the reason, it should be presented up front where auth are discussing the potential weaknesses of the 833/cm denominator. Moreover, it is not clear to me that the 833-960/cm spectral difference in the water vapor continuum is sufficiently substantial to provide the aerosol clarification needed. Because the continuum is invoked, it would be important to provide more detail or citations to make the point.

We will change the order of arguments. Spectral changes in water vapor absorption can be seen either in figure Fig. 3.1 in Goody and Yung, 1989 or Fig. 7.6 in Petty, 2006.

On P4386 auth present Figure 2 to illustrate the CI/AI differences. I had a very difficult time understanding the figure and attendant discussion. On L8 auth state that "For regions with CI<2 there are certainly ice clouds…" To what do they attribute this certainty? They seem to be presuming that CI<2 is proof of ice. If this is the case, it is illogical given that the purpose here is to assess the accuracy of CI and AI against some objective truth (cirrus, other clouds, volcanic sulfate, and other aerosols in this post-Nabro orbit). It would be more appropriate here to present some independent evidence of clouds, aerosols, clear sky with which to assess MIPAS UTLS signals. The reader has no information on the true curtain of particles here. A similar presumption based on MIPAS indices is in L11, "For the AI we see that in clear air regions the AI remains above 7." Again, the reader does not know the truth about these scenes (i.e. whether or not that region really is clear) and presumably the intent should be to judge how truthful/accurate the CI and AI are with respect to independent information. On L15, following the discussion of Figure 2 (1 orbit of MIPAS data), auth state that an AI threshold of 7 was discerned based on a visual inspection of all MIPAS orbits in 2011. They present no objective basis on which to assess aerosol/clear-sky boundaries in 2011. In my opinion, the method described here is vague and unsupportable.

Figure 2 was difficult for me to interpret. Even though latitudes and longitudes are given, I would suggest a map panel be presented so the reader can easily see where in the world this orbit was. The color difference between important index thresholds (e.g. AI=7) was not stark enough for easy feature discernment. Moreover, there is no tropopause information on the plot, hence it is difficult to assess the UTLS. A tropopause line would help this figure enormously. Auth point out some features in Figure 2 that I could not unambiguously identify. For instance the "detached layer" (L13) is murky to my eye. Perhaps auth could annotate the figure with arrows or some such device to make these specific features readily identifiable.

P4386, 2nd paragraph. The discussion of the compromised behavior of AI above 25 km is important but is not supported well. Auth refer to Figure 1 while discussing artifacts above ~25 km, which is the top of Fig. 1. I.e. it's difficult to know what the reader is supposed to see in Fig. 1 to understand the issue. Please either clarify the discussion or increase the altitude range of Figure 1.

P4387, L5. Auth state "To further confirm the ACI threshold of 7 we derived…" In my assessment, the ACI=7 wasn't confirmed (see my comments above), so a further confirmation is not possible. Perhaps auth mean "evaluate" instead of "further confirm"?

Here we should change the line of argumentation. It would be better to start with the simulations (including figures illustrating the differences of single clear air CI and ACI profiles) and then go on to the observations.

P4387, L5-26. Here auth use a radiative transfer model with a module (MT_CKD) that is considered to be inaccurate for real atmospheric conditions to assess their empirical AI=7 clear/aerosol threshold. The simulations are grossly at odds with the MIPAS data. Auth then conclude that their theoretical approach has uncertain merit, so fall back exclusively on the empirical approach's result. This exercise does not, I my assessment, "further confirm" the ACI threshold. It's not clear to me what good this particular simulation exercise was. I do believe that something in addition to the empirical/visual thresholding is called for. I'd suggest auth either reformulate the RTM strategy or invoke independent aerosol/cloud observations to evaluate the

ACI.

We did not intend say that the radiative transfer simulations are useless or wrong. They actually perform very well for the vast majority of the MIPAS measurements from 2005-2012. Only for low altitudes between 5-10km in the tropics (that were covered by MIPAS measurements only between 2002-2004) there are issues with the water vapour continuum in the simulations. Will be rephrased to get this point across more clearly.

Section 3.2: I describe below the details of my overall major concern with this section. Some of the items within this description can be considered minor but they are all together to make my point.

P4388, L3. Auth begin section 3.2.1 discussing their choice of windows "for the discrimination between aerosol and ice clouds." It seems to me that they had already done that in section 3.1. Hence the introduction to this discussion seems to need clarification. On L10 they claim to have identified in Section 3.1 three window regions, yet there is no such attempt in 3.1. Moreover, of the three bands listed, only one (960-961/cm) has roots in 3.1. This makes me wonder if indeed there is material they intended for the AI/CI definition that was left out of 3.1. Please clarify.

We used the same methodology as described in section 3.1 also in sect 3.2. Will be clarified.

On P4388 auth expressly limit the aerosol typing that they intend to model and discern as volcanic ash and sulfate. It is here specifically that I would expect auth to either include the full suite of aerosols that MIPAS encounters in the upper troposphere, or to explain why they are limiting the scope to ash and sulfates. If they intend to explore the more representative array or aerosols in future work, this would be the place to make that point.

We did not intend to classify the whole aerosol suite. Please see comments above.

P4389, L11-14. I was confused about these various ranges of numbers and the meaning thereof. It seemed to me that there was a lot of overlap, making it hard to draw any conclusions. I ask auth to provide clarification on how they interpret these ranges of percents.

Please see Fig 3d. Ranges indicate the changes over the considered spectral range / between the 3 windows. Will be added to the text.

P4389, 3.2.2. On L23 auth discuss how they defined "regions where they expected" to find four cloud/aerosol scenarios. They do that by listing broad latitude ranges in Figure 4, but no additional qualification. E.g. there is no longitudinal information that might be logical if they are focusing on a volcanic plume. They give no altitude range selection criteria. In my assessment, this gives too little help to the reader. And if indeed there are no other criteria than in the Figure caption, it seems too broad a selection scope. Hence interpreting the patterns in Figure 4 is met with great uncertainty as to just what the true physical constituents are that are in the plot. One suggestion I have is to segregate data points by their tropopause-relative position. The reader would benefit greatly if he/she could see at a glance where the tropospheric/stratospheric particles and ice are.

In the revised manuscript we will describe the selection criteria in more detail. Time: before/after a known volcanic eruption event. Altitude range: 25km – MIPAS lowest tangent altitude (approx. 6-10km depending on latitude). Latitude range: as given in the figure caption, mainly to constrain the hemisphere and to exclude/include PSCs. Longitude range: -180-180, as we had only the information that there was/was not a volcanic eruption somewhere in the hemisphere. Our expectation was, if there is some other particle type than ice, it should stand out from the non-perturbed scenario in Fig 4a.

We will also consider changing the line of argumentation, to start with the simulations and then go on with the observations.

In the above comments I have mentioned my concern for the lack of an independent aerosol/cloud observation data set with which to compare the MIPAS indices. It seems to me that the CALIPSO vertical feature mask, and even CloudSat data, could be put to great use in this application. The time coincidence is not ideal, but for the types of data presented herein (e.g. the global MIPAS curtain 2 months after Nabro) time coincidence is not a strict criterion.

We thought that too, however, concerning the comparison with Figure 2, by mid-August the browse image CALIPSO vertical feature mask does not indicate the Nabro aerosol layer anymore. Only in the nighttime backscatter signal the layer is still visible. Hence, we initiated a comparison of MIPAS sulfate aerosol detections with the CALIOP nighttime aerosol product (provided by J.-P. Vernier) for the Nabro eruption between June to early August 2011 from 0-50N, ground based lidar measurements 50-68N between August 2011- February 2012, and ground based twilight measurements in August 2011. Since this comparison turned out to be anything but a trivial task and is quite extensive, we decided to present it in a separate paper (which we mentioned on page 4392 line 24).
Regarding a comparison of MIPAS and CALIPSO curtains, previous studies found that for ice clouds at low and mid-latitudes the match times are not close enough (Hurley et al., 2011). Hence, ice cloud comparisons would only feasible on a statistical basis. We think the same holds for CloudSat (also in the A-train).

Minor

In the introduction, auth survey prior attempts to retrieve UTLS cloud/aerosol types and promote the advantages of IR limb sensing. No mention is made of the work done with HIRDLS. Sembhi et al. (2012) is cited but not for its use of HIRDLS cloud detections.

Although HIRDLS is not a hyperspectral instrument we agree that it should be included in the discussion since the older IR instruments ISAMS and CLAES are also mentioned.

Also in the survey of prior vertically resolved aerosol/cloud retrievals, auth use the term “limb measurements in the IR” or variants thereof, and limit their survey to measurements of IR emission. If this is their intent, they should specify “emission.” Otherwise, they leave out several important NIR- and IR-based accomplishments, e.g. HALOE and SAGE (Thomason, 2012).

Our focus is on IR emission instruments.

P4385, L7. Why is the CI cloudy-air threshold expressed as a range? What is the significance of CI-1.8?

According to Sembhi et al., 2012 the CI threshold depends on altitude, latitude and season. CI 1.8 is generally used for cloud filtering before trace gase retrievals e.g. by ESA. Will be clarified.

P4389, L22. Auth mention “four selected days” and then “(about 14 orbits).” There are ~14 orbits in a single day. So does this mean 14 orbits spread over 4 days? Please clarify and give the dates.

In Fig. 4 each panel is for a single day and comprises 14 orbits. (14 orbits per day). Will be clarified.

Figure 4 caption. This is presumably a northern summer period, hence “PSCs (0-90N)” should be 0-

90S.

The data in Fig 4d were measured on 29. January 2011, which is northern hemisphere winter. (We suppose that during copy editing the vertical space between the top and bottom row got lost and the x-axis label of the upper panel and the title of the lower panel are too close now. Will be improved.)

Figure 4 caption. “All figures comprise …single day.” This is inconsistent with the text “four selected days.”

All panels comprise a single day, which are about 14 orbits.

Figure 6. The height-scale colors provide too little contrast between height bins. Please consider a clearer height differentiation.

This will be changed.

References

Clarisse, L., M. Fromm, Y. Ngadi, L. Emmons, C. Clerbaux, D. Hurtmans, and P.-F. Coheur (2011), Intercontinental transport of anthropogenic sulfur dioxide and other pollutants: An infrared remote sensing case study, Geophys. Res. Lett., 38, L19806, doi:10.1029/2011GL048976.

Cottle, P., Strawbridge, K., McKendry, I., O'Neill, N., and Saha, A.: A pervasive and persistent Asian dust event over North America during spring 2010: lidar and sunphotometer observations, Atmos. Chem. Phys., 13, 4515-4527, doi:10.5194/acp-13-4515-2013, 2013.

Fromm, M., et al. (2008), Stratospheric impact of the Chisholm pyrocumulonimbus eruption: 2. Vertical profile perspective, J. Geophys. Res., 113, D08203, doi:10.1029/2007JD009147.

Fromm, M., D. T. Lindsey, R. Servranckx, G. Yue, T. Trickl, R. Sica, P. Doucet, and S. Godin-Beekmann (2010), The Untold Story of Pyrocumulonimbus. Bull. Amer. Meteor. Soc., 91, 1193–1209, doi: http://dx.doi.org/10.1175/2010BAMS3004.1

Husar, R. B., et al. (2001), Asian dust events of April 1998, J. Geophys. Res., 106(D16), 18317–18330, doi:10.1029/2000JD900788

Liu et al. (2013), Transpacific transport and evolution of the optical properties of Asian dust, Journal of Quantitative Spectroscopy & Radiative Transfer 116 (2013) 24–33.

Solomon, S., Daniel, J. S., Neely, R. R., Vernier, J. P., Dutton, E. G., & Thomason, L. W. (2011). The Persistently Variable “Background” Stratospheric Aerosol Layer and Global Climate Change. Science. doi:10.1126/science.1206027 Thomason, L. W. (2012), Toward a combined SAGE II-HALOE aerosol climatology: an evaluation of HALOE version 19 stratospheric aerosol extinction coefficient observations, Atmos. Chem. Phys., 12, 8177-8188, doi:10.5194/acp-12-8177-2012.

Vernier, J.P., T. D. Fairlie, J. J. Murray, A. Tupper, C. Trepte, D. Winker, J. Pelon, A. Garnier, J. Jumelet, M. Pavolonis, A. H. Omar, and K. A. Powell, 2013: An Advanced System to Monitor the 3D Structure of Diffuse Volcanic Ash Clouds. J. Appl. Meteor. Climatol., 52, 2125–2138. doi: http://dx.doi.org/10.1175/JAMC-D-12-0279.1

References in addition to references in manuscript

Goody, R. M. and Yung, Y. L.: Atmospheric Radiation, Theoretical Basis, Oxford University Press, 1989

Höpfner, M.: Study on the impact of polar stratospheric clouds on high resolution mid-IR limb emission spectra, J. Quant. Spectrosc. Radiat. Transfer, 83, 93–107, 2004.

Höpfner, M., Larsen, N., Spang, R., Luo, B. P., Ma, J., Svendsen, S. H., Eckermann, S. D., Knudsen, B., Massoli, P., Cairo, F., Stiller, G., Von Clarmann, T., and Fischer, H.: MIPAS detects Antarctic stratospheric belt of NAT PSCs caused by mountain waves, Atmos. Chem. Phys., 6, 1221–1230, 2006.

Hurley, J., Dudhia, A., and Grainger, R. G.: Retrieval of macrophysical cloud parameters from MIPAS: algorithm description, Atmos. Meas. Tech., 4, 683–704, 2011

Keckhut, P., Hauchecorne, A., Bekki, S., Colette, A., David, C., and Jumelet, J.: Indications of thin cirrus clouds in the stratosphere at mid-latitudes, Atmos. Chem. Phys., 5, 3407–3414, 2005.

Petty, G.W.: A first Course in Atmospheric Radiation, Sundog Publishing, 2006.

Spang, R., Günther, G., Riese, M., Hoffmann, L., Müller, R., and Griessbach, S.: Satellite observations of cirrus clouds in the Northern Hemisphere lowermost stratosphere, Atmospheric Chemistry and Physics, 15, 927–950, doi:10.5194/acp-15-927- 2015, http://www.atmos-chem-phys.net/15/927/2015/, 2015.