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Interactive comment on "Stratospheric and tropospheric NO_2 variability on the diurnal and annual scale: a combined retrieval from ENVISAT/SCIAMACHY and solar FTIR at the Permanent Ground-Truthing Facility Zugspitze/Garmisch" *by* R. Sussmann et al.

Anonymous Referee 1

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General comments

The manuscript tries to adress the accuracy of SCIAMACHY NO2 retrievals by inter-comparing mountain-based FTIR observations with SCIAMACHY data gathered within 200 km of the mountain station. The introduction of the "virtual coincidence" and "pollution-clearing scheme" are useful and good ideas for validation. However, this

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reviewer has learned very little on whether the Bremen SCIAMACHY NO2 data can now be considered validated. Qualitatively, SCIA and FTIR appear to be in agreement (Fig. 7), but there is a large (seasonally varying) difference between the two, and the cause of this should be unravelled. Obviously, SCIA retrievals include contributions from the troposphere, and this likely explains an important part of the difference. This leaves us still with the question of SCIA NO2 accuracy with respect to errors in the NO2 absorption cross-section, the possible offset introduced by using a reference spectrum recorded on 15 december 2002, the empirical correction (1.0E+15 molec.cm-2), and other retrieval errors such as introducing stratospheric aerosols in AMF calculations, not taking into account clouds, etc. The authors could at least try to discriminate retrieval errors from sensitivity differences (plotted together in Fig. 7) by focusing on SCIA pixels over Zugspitze in 'clean' situations or in situations with moderately high clouds, when sensitivity to tropospheric NO2 will be reduced.

A second goal of the manuscript is to present a new method for a combined ground/satellite-based retrieval to estimate tropospheric NO2. The idea is to use the different sensitivities of the FTIR (mainly sensitive to stratospheric NO2), and the satellite measurements (sensitive to both tropospheric and stratospheric NO2), assuming some vertical distribution of NO2, to arrive at an estimate for the tropospheric NO2 column. This is a very good idea, and should certainly be published. The development of the idea however leaves much to be wished for. First of all, there is very little discussion of the quantitative meaning of the result. This is related to the introduction of the retrieval constraint (equation (4)). This constraint is a reasonable starting point for the method, but in reality there are strong deviations from the proposed profile shape (Fig. 10), notably in the troposphere. Moreover, the fixed kernel for SCIA is hopelessly unrealistic. Kernel shapes are very sensitive to forward model parameters such as clouds and albedo. In other words, because clouds, albedo, and profile shape are so variable in time and space, Fig. 12(c) is unlikely to give a realistic view of the 'clean background tropospheric column'. In my opinion, it would be best to recompute the 'clean background column' taking into account all relevant forward model parameters.

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If this is not possible at this stage, I would strongly suggest that the result -Fig 12(c)- is at least discussed in terms of validity given the objections raised above.

Specific comments page 2378, line 8: remove 'selection'

p 2378, l11: what does 'uncorrected' mean here?

p2379, l5: remove 'respectively'

p2379, l13-15: from surface measurements and trajectory calculations it was well-known that pollution events could last long and also be tranported. I would like to see a justification for the statement that 'Such pollution events had been underestimated...'

p2379, l19: I only see one approximation, the TEM by Richter and Burrows. However there are more: the Richter et al. method to scale a CTM stratosphere, and data-assimilation of NO2 slant columns into a CTM (Boersma et al., JGR, 2004) deserve to be mentioned. And there are methods to account for strotospheric variability by employing filtering techniques on the data alone (Leue et al., 2001).

p2380, l1: On the other hand, work by Heland et al. (GRL, 2002) and Martin et al. (JGR, 2004) has given clues that NO2 satellite is not so bad. Please also mention this.

p2380, l6: 'in general terms', what does this mean?

p2385, l14-16: I think an error propagation study is in order here. Later on, the results of such an analysis could be compared with the results in section 2.9.

section 2.7: Nice concept. However, more words should be spent on the error in the estimated FTIR column based on application of the daytime increasing rate.

p2386, l5: why is Fig.7 introduced before Figs. 5 and 6?

p2386, l8-9: (i) in terms of validation, this is a bad idea. Comparing FTIR and SCIA on days that SCIA did not measure will result in checking the functional fit to the SCIA data and this is only an indirect way of validation.

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p2386, l9-12: (ii) here a similar reasoning holds. I agree that an idea can be obtained on the day-to-day scatter, but then the precision and accuracy of the functional fit should be very high.

p2387, l19: why are there brackets around the pixel size?

p2388, l8: 'no cloud clearing'. This is a fundamental weakness of the manuscript. First of all, cloud information may help the authors: the pollution clearing scheme might well be simplified when cloud information is used. Second, not taking into account the effects of clouds introduces problems in interpreting the results: cloudy pixels generally screen the NO2 pollution underneath and strongly influence the sensitivity of any NO2 above (or within) the cloud. Even in the stratosphere, the sensitivity to NO2 is significantly enhanced when clouds are present. This effect should be accounted for in the AMF. It may well explain part of the bias of SCIA relative to FTIR.

p2388, l13-14: This line is nonsense. The result of DOAS is NOT that slant column densities are integrated along the effective light path, but rather that slant column densities should be interpreted as the column of molecules along the effective light path.

p2388, l21: it is new to me that the AMF is dependent on pressure profiles or do the authors mean surface pressure? Moreover, it is a bit odd not to mention the most important forward model parameter for AMF calculations here, i.e. clouds.

p2389, l2: what is the motivation or justification for assuming stratospheric background aerosols?

p2390, l17: I fail to see why days with $>$ 12 measurements would avoid including polluted data.

p2392, l1: I understand the point, but there are subtle issues here. Over the Pacific, a 'reduced US standard NO2 profile' is likely a much better assumption for AMF calculations than over the Zugspitze area. Assuming the 'reduced' profile will result in higher AMF values and therefore reduced scatter. If a non-reduced profile would be

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used in the AMF, the scatter might well exceed 10%, and hence corrupt the validity of the pollution-clearing approach.

p2393, l17-18: I don't agree. See general comments.The assumption of identical sampling characteristics is just wrong, as the authors are well aware of. Therefore, an effort should be made to separate the different sensitivities from retrieval errors, as suggested above.

p2394, l21: it might be instructive for readers to learn why the FTIR retrieval is not senith-angle dependent.

p2397, l7: I think the numbers mentioned here and in Table 3 should be discussed in terms of validity. They are true given assumptions on albedo, clouds, etc.

p2398, l23, Fig. 12b is introduced before Fig 11 and 12(a).

p2399, l2, why is the vector x_trop defined following equation (5)? I see the need to translate mixing ratios to subcolumns, but multiplying VMR with the FTIR AMF's is introducing sensitivities in a profile that is supposed to serve as an a priori profile, subject to the kernels. Therefore, applying AMF's seems incorrect to me here or am I missing something?

p2399, l2: what is the sensitivity of the results for different assumptions on the vector x_trop? See general issues.

p2399, l12: add Eq. (2).

p2401, l8-9: were these column values observed by ground-based instruments or by GOME, as suggested on p2404, l10-11. If GOME, than the 'evidence' comes down to reasoning that both SCIA and GOME observe NO2 in the same range -in completely different areas- and that therefore SCIA is in a 'reasonable' range. The statement in l10-11 would then be meaningless since we don't know if we can trust GOME over the Po Valley.

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p2401, l1: lifetime also plays a role here as NOx has more loss pathways in summer than in winter. It is not just the emissions.

p2402, l25-27: I am lost here. I don't see the fact that the SCIA-true difference is smaller than the FTIR-true difference gives the proof. Hopefully the authors can explain it more clearly.

p2403,l20: what does 'uncorrected' mean here?

p2403, l28: 'true to a best approximation': isn't this the same as stating that you think -for some reason- that this IS the best approximation. And if so, why?

p2404,l14-15: this line is illustrative of what is missing from the manuscript. It is a simple statement without any proof from the results shown in the manuscript. It is certain that there are SCIA errors, and the whole idea of a validation study is to quantify these errors, and to provide at least a guess of where these errors come from, and how they could be eliminated. None of this is dealt with in the manuscript. Even the statement that the errors will not exceed the clean tropospheric column values may be false. The clean tropospheric column may well be underestimated due to kernel and profile shape issues mentioned above, and if this were the case, the errors in SCIA would exceed the reported 'clean tropospheric columns'.

p2404, l19: in principle yes, but it is not sufficient to use one SCIA kernel and one 'retrieval constraint' as is done in the manuscript. Every single SCIA observation then needs to take into account the best estimate of the forward model parameters. This should be mentioned.

p2404, last section. The manuscript would benefit from a rough sketch on the feasibility of an 'integrated observing system'. How many appropriate sites are available at the moment? Is their geographical distribution good enough?

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 2377, 2005.