SRON
Netherlands Institute for Space Research

FINAL REPORT

Project number: GO 2004/34

Project title: Tropospheric Ozone Re-Analysis (TORA)

Researcher(s): Dr. A.T.J. de Laat, Dr. R.J. van der A

Supervisor(s): Dr. M. van Weele

Starting date: 1-2-2006

Duration of project: 36 months (24 person months, 100% first year, 50% second and third year)

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1. *Initial aim/research problem(s):*

The goal of the TORA project is to create a global tropospheric ozone climatology based on measurements by the GOME instrument, for the period 1996-2001. It is estimated that the accuracy of the climatology will be sufficiently high to make it valuable for the study of the inter-annual and seasonal variability and the regional differences in tropospheric ozone and related radiative forcing.
The climatology will be created by subtracting the stratospheric part from the total ozone column, where the stratospheric part is obtained from assimilation of GOME ozone profiles into an atmospheric chemistry model. Theoretical error estimates will be provided. Ozone sondes will be used for the validation of the absolute columns and the surface-to-tropopause mean mixing ratios and associated uncertainties.

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2. *Summary of the course of the project (max. 2 x A4):*

The TORA project consisted of six work packages.

For WP1 GOME O_3 profiles and total columns were produced according to plan. Halfway the project, a reprocessing of the GOME O_3 profiles was performed using an improved version of the retrieval algorithm. The reprocessed O_3 profile data was used in the latter half of the project.

For WP2 the GOME O_3 profiles were assimilated in the TM model. The assimilation code that was planned to be used was based on an older version of the TM model (TM3). This model version was going to be replaced by a new version of the model (TM5), which has gradually been introduced in the TM modelling community whereas the older version (TM3) was gradually phased out and not supported anymore. Therefore, the assimilation module had to be implemented in TM5, a non-trivial exercise given the different model structures of TM3 and TM5. In addition, the data format of GOME O_3 profiles available for TORA had changed compared to the data used in the TM3 assimilation version, which required additional modifications to TM5. The implementation of the assimilation in TM5 (including enabling TM5 to read the new data format) was performed within WP2, but it was not scheduled within the TORA project. This took a considerable amount of time – in no small part related to the fact that personnel responsible for technical support had by then left KNMI and as such were no longer available for support. Furthermore, initially the assimilation turned out to be unstable, producing unrealistic numbers and crashes of simulation runs. Additional effort had to be put in finding a solution for this fundamental problem. All in all, these unforeseen circumstances caused a considerable delay within the project, and it was decided halfway the project to request to spread the second half of the project (one year) over two years for 50% of the time of the researcher. This request was graciously granted by GO and has helped the progress of TORA tremendously since there was a considerable amount of ‘idle’ time within the project due to, for example, the long computational time required for running the assimilation model for one or more years (six year assimilation took about 3 months). This time could be spend on other projects without hampering the progress of TORA.

In WP3 the assimilation results were used for applying the TORA method – subtracting the assimilated stratospheric O_3 column from total columns observations – and delivering a tropospheric O_3 column. In WP3 there was an additional task to investigate the vertical sensitivity of the total O_3 column. This task was not performed because of the unexpected results outlined below.

In WP4 the TORA product was to be analyzed by comparing the assimilated stratospheric O_3 columns with independent observations and by comparing tropospheric O_3 columns with independent observations and full chemistry-transport model results. This task was performed for one year of data (2000). From WP3 a number of errors and uncertainties were identified and analyzed, and it was concluded that for rather fundamental reasons the assimilation product was not up to standards – i.e. useful to deliver valuable tropospheric O_3 columns outside the tropical band.

Within WP5, the tropospheric O_3 columns were to be post-processed into a data format useful for a broad research community and further analyzed. The assimilation of GOME O_3 profiles and application of the TORA method was performed for the period 1996-2001 as planned. However, as a consequence of findings in WP4 there was no incentive to put much effort in post-processing all the data, and only part of the post-processing was performed. Rather, additional effort was put into further analyzing the errors and uncertainties discovered in WP4 and finding ways to improve the method outside the tropical band with a focus on the extratropical UTLS. One important finding from WP4 and WP5 was that higher-resolved observational data – O_3 profiles – were essential for useful results. As a consequence, some effort was put into analyzing new O_3 profile data from the GOME-2 instrument flying on METOP and investigating whether the GOME-2 O_3 profiles were of better quality. The analysis of the GOME-2 data suggests that GOME-2 indeed meets the required data quality, and therefore might be better suited for the TORA methodology on a global scale. Since this was the first ever analysis of GOME-2 O_3 profile data, the results from this analysis were written down in a separate paper [van Peet et al., 2009].

Finally, WP6 was meant for archiving and publicly disseminating the data and publishing results. Due to the sub-standard quality of the data, there was not incentive to disseminate the data. The results of the TORA project nevertheless are very useful for the research community and have been written down in a single paper [de Laat et al., 2009].

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3. *Tangible results (max. 2 x A4):*

This research project has developed a six year dataset of assimilated GOME O_3 profile measurements and six years of residual tropospheric O_3 columns from subtracting the assimilated stratospheric O_3 column from total column observations, the so-called TORA method.

The comparison of model simulations without assimilation for the UTLS region (O_3 column between 250 and 50 hPa) with independent O_3 sonde observations shows that the free-running TM5 model with the CARIOLLE linearized O_3 chemistry scheme produces realistic tropospheric and lower stratospheric O_3 variations, both on short day-to-day as well as longer seasonal timescales, although for the tropics some tropospheric biases could be detected. The agreement improves for monthly and climatological means. The results also show that the RMS differences are quite large, especially for middle and high latitudes.

When assimilating GOME O_3 profiles in the TM5 model simulation the UTLS results in general do not improve. Correlations and RMS differences outside of the tropics degrade albeit not very significantly. Within the tropics correlations and RMS difference improve slightly. On the other hand, biases improve somewhat for the assimilated UTLS columns. The comparison of model results for the TOC shows that overall the correlations and RMS differences degraded when assimilating GOME O_3 profiles, although on the other hand the biases improved. This behaviour can be understood by noting that Chemistry-Transport Models (CTM) generally produce a too strong stratospheric transport, leading to accumulation of O_3 in the stratosphere at mid-latitudes. This is also the case for the TM5 model used in this study, as reported in de Laat et al. [2007]. Figure 1 shows six years of annual mean differences in UTLS O_3 columns (1996-2001) between a control simulation and a simulation with assimilation of GOME O_3 profile measurements. The difference field shows a gradual increase over the years, showing that the assimilation keeps modelled UTLS O_3 "in check", i.e. the bias by accumulation of O_3 due to the overstrong stratospheric circulation in the model cannot manifest itself because of the continuous assimilation of O_3 profiles.

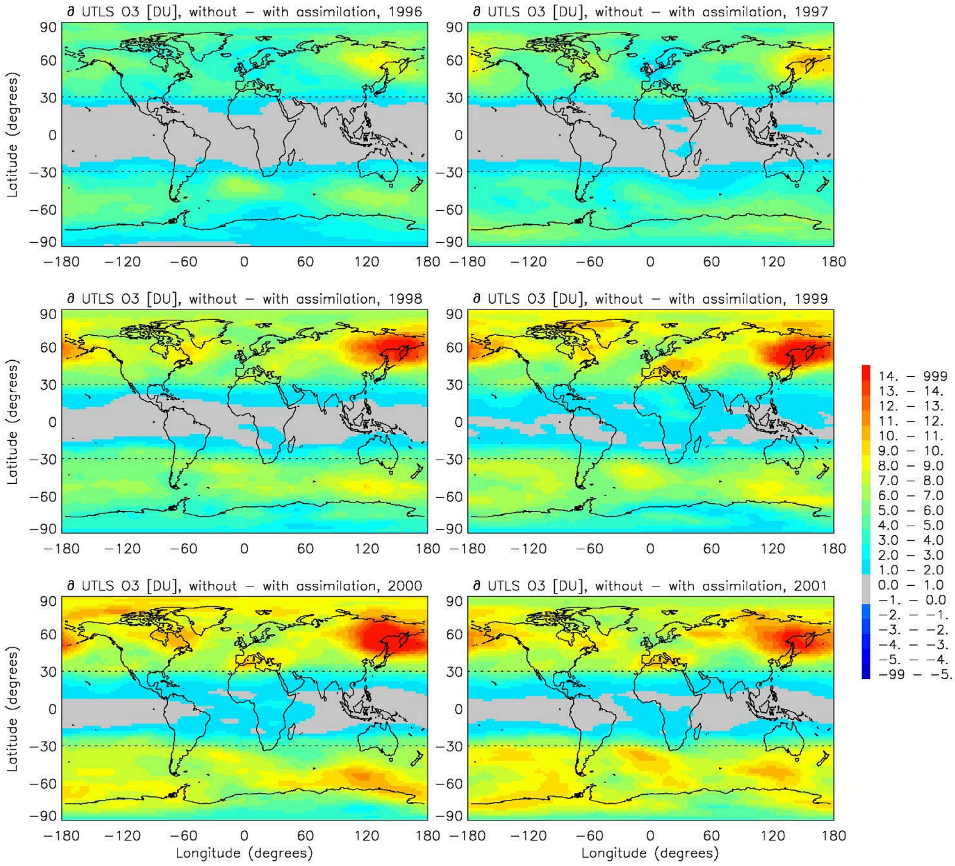

Figure 1. *Annual mean UTLS (250-50 hPa) O_3 column differences [DU] of the free running TM5 model simulation and the TM5 GOME O_3 profile assimilation run for every single year between 1996 and 2001. Values between -1 and +1 DU are indicated in grey.*

Summarizing the evaluation of model results we conclude that for GOME O_3 profiles the assimilation does not improve the comparison with independent sonde observations. The main reason is likely the very large footprint of the GOME pixel (960×80 km), which is larger than the typical correlation length of upper tropospheric (L ~ 100 km) and lower stratospheric (L ~ 300-400 km) O_3 variations. Hence, the assimilation probably introduces errors as much as it improves the representation of UTLS O_3. Additional errors may be introduced by allowing up to 90% cloud covered GOME pixels in the assimilation, so that it becomes unclear where exactly the troposphere is sampled. Due to the vertical smoothing of the GOME O_3 profiles this error is also transferred to the lower stratosphere in the GOME O_3 profile. Additionally, instrumental and retrieval errors may have also contributed, although this was not analyzed given the current status of the evaluation presented here.

Applying the TORA method to derive tropospheric residuals shows that outside of the tropics the day-to-day variations in the residuals are much larger than the observed variability – even yielding negative residuals. On the other hand, the average bias is smaller in the residuals compared to the TOC directly from the assimilation, and within the tropics the residuals appear realistic, suggesting that the assimilation of GOME O_3 profiles do not degrade the tropospheric residuals. Figure 2 shows two examples of comparison of the TORA residuals with O_3 sonde observations for a mid-latitude and tropical location, highlighting these findings.

The spatial distribution of annual mean residual columns shows some apparent realistic variations –especially in the tropics. Given the previous validation with sonde observations it is not very likely that the spatial variations in the annual means and the interannual variability contain valuable information outside of the tropics.

Nevertheless, the results also clearly point in the direction where to improve this methodology. First of all, the spatial resolution of the model must be improved to at least 1°×1°, preferably better, so that UTLS O_3 variations can be realistically simulated. Realistically means that the model resolution should be smaller than the empirically established correlation length of UTLS variations, which is about 100 km or 1° according for the upper troposphere according to Sparling et al [2006]. In addition, the observations that are assimilated should also have a footprint that is at least of similar size, preferably smaller.

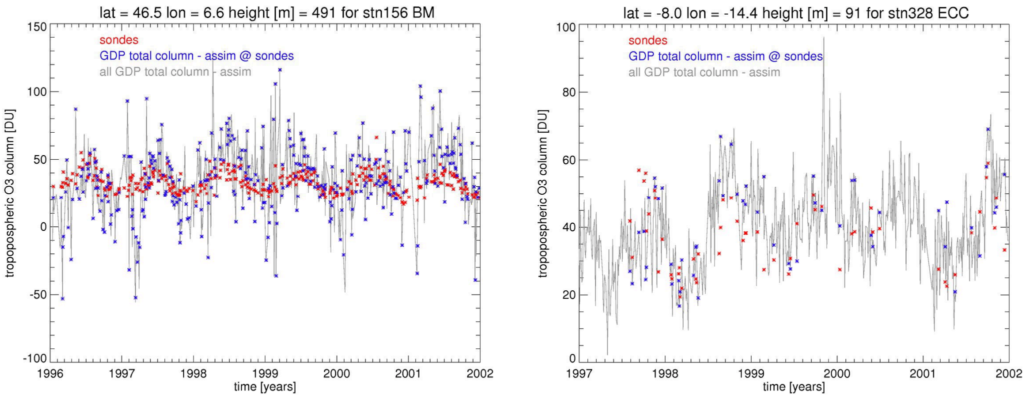

***Figure 2**. Comparison of GDP v4.1 GOME total O_3 columns minus the TM5 assimilated stratospheric O_3 columns and WOUDC O_3 sondes. The left panel is for mid-latitude station Payerne, Switzerland (stn156), the right panel for tropical station Samoa (stn328). Shown are the individual observed (red) and total-minus-assimilation columns (blue: GOME GDP collocated with sondes; grey: all GOME GDP columns).*

Schoeberl et al. [2007] presented results for a similar-but-different methodology, similar in a sense that the method also used a sort of assimilation of O_3 profiles, but different in sense that a different assimilation approach and different observations were used (O_3 profiles from the Microwave Limb Sounder). Nevertheless, with a better spatial resolution and using MLS stratospheric O_3 profiles – which have a much better vertical resolution – apparent more realistic residuals were derived and they could observe realistic O_3 variations on smaller spatio-temporal scales (roughly 1°×1° resolution for a five-day average).

Important for the TORA methodology is that considerable improvement in O_3 profile quality and accuracy has been reported for GOME-2 O_3 profiles compared to GOME O_3 profiles, while at the same time GOME-2 O_3 profiles also have the necessary smaller footprints [van Peet et al., 2009; submitted for publication]. Future tests may also use different observations, like the aforementioned MLS or other limb observations, for example from SCIAMACHY or MIPAS or OMI O_3 profiles, the latter having an even smaller footprint than GOME-2.

One drawback for the use of for example GOME-2 is that the number of observations that need to be assimilated increases considerably. The assimilation step is by far the time limiting step in the TORA methodology, hence increasing the number of observations to be assimilated increases the computational time accordingly. However, the results from Schoeberl et al [2007] suggest that realistic results might be achieved by using only a subset of all available O_3 profile observations, as the spatial coverage of MLS is considerably less that of GOME-2 or OMI and nevertheless realistic results were reported.

Summarizing we conclude that the TORA methodology provides realistic tropospheric O_3 columns for tropical latitudes. Outside of the tropics, accurately estimating the tropospheric O_3 column is hampered by the too large ground pixel of the GOME O_3 profile observations and possible insufficient accuracy of the O_3 profile observations as well as a too large horizontal resolution of the TM5 assimilation model. Recommendations for future application of the TORA methodology are to use O_3 profile observations with a smaller spatial footprint – preferably 1°×1° degrees or smaller – and a better horizontal resolution of the assimilation model – also preferably 1°×1° degrees or smaller. Fortunately, UV-VIS O_3 profile observations with the required spatial footprint are becoming available from the GOME-2 and OMI instruments, which in the near future will be used for applying TORA methodology.

The results of this research project have been written down in a scientific paper that has recently been submitted to ACP [de Laat et al., 2009].

References

de Laat, A.T.J., J. Landgraf, I. Aben, O. Hasekamp, and B. Bregman, Validation of Global Ozone Monitoring Experiment ozone profiles and evaluation of stratospheric transport in a global chemistry transport model, J. Geophys. Res., 112, D05301, doi:10.1029/2005JD006789, 2007.

de Laat, A.T.J., R.J. van der A and M. van Weele, Evaluation of tropospheric ozone columns derived from assimilated GOME ozone profile observations, submitted to ACP, 2009.

van Peet, J.C.A., R.J. van der A., A.T.J. de Laat and O. Tuinder, The height resolved ozone hole structure as seen by GOME-2, submitted to Geophys. Res. Lett., 2009.

Schoeberl, M.R., J.R. Ziemke, B. Bojkov, N. Livesey, B. Duncan, S. Strahan, L. Froidevaux, S. Kulawik, P.K. Bhartia, S. Chandra, P.F. Levelt, J.C. Witte, A.M. Thompson, E. Cuevas, A. REdondas, D.W. Tarasick, J. Davies, G. Bodeker, G. Hansen, B.J. Johnson, S.J. Oltmans, H. Vömel, M. Allaart, H. Kelder, M. Newchurch, S. Godin-Beekman, G. Ancellet, H. Claude, S.B. Andersen, E. Kyrö, M. Parrondos, M. Yela, G. Zablocki, D. Moore, H. Dier, P. von der Gathen, P. Viatte, R. Stübi, B. Calpini, P. Skrivankova, V. Dorokhov, H. de backer, F.J. Schmidlin, G. Coetzee, M. Fujiwara, V. Thouret, F. Posny, G. Morris, J. Merrill, C.P. Leong, G. Koenig-Langlo and E. Joseph, A trajectory-based estimate of the tropospheric ozone column using the residual method, *J. Geophys. Res., 112*, doi: 10.1029/2007JD008773, 2007.

Sparling, L.C., J.C. Wei and L.M. Avallone, Estimating the impact of small-scale variability in satellite measurement validation, J. Geophys. Res., 111, doi: 10.1029/2005JD006943, 2006.

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4. *Have the original research aims been achieved or adjusted? Has the project served other purposes in addition?*

The original research aim of the project – *i.e.* reconstructing a six year tropospheric O_3 climatology has been achieved. However, one of the important conclusions of the project is that the observational GOME data used for the assimilation has insufficient spatial resolution to provide useful data outside of the tropics. Hence, a global six year O_3 climatology for scientific was not produced for the simple reason that it was of insufficient quality. However, the findings of this project also clearly point into the direction where improvement can be expected: higher resolved O_3 profile observations, which are becoming available from for example GOME-2 or OMI, and an assimilation model with a better horizontal resolution, which is just a matter of available computational power.

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5. *Has the project generated new scientific questions? If so, specify:*

One particular result of TORA was the search for better satellite O_3 profile observations. Therefore, GOME-2 O_3 profile data (see question 2 of this report) was analyzed for its quality. As a result, it was discovered that quality the GOME-2 O_3 profile observations had significantly improved compared to the GOME O_3 profile observations. For example, it is only with GOME-2 that it is possible to observe the complete vertical distribution of O_3 from the surface all the way to the upper stratosphere on a day-to-day basis, including for example the vertical and horizontal structure of the Antarctic ozone hole. This is a unique achievement that cannot be delivered by any other satellite remote sensing instrument or facility. This finding has lead to a number of new scientific questions, for example with regard to our current estimates of the size of the Antarctic ozone hole, which we may have underestimated.

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6. *Are the results achieved of relevance to current research by others? If so, specify:*

Yes, the findings of the project have resulted in clear recommendations for future work. The TORA method can be improved upon by using better (stratospheric) O_3 profile observations and by increasing the horizontal resolution of the assimilation model. The quest for better stratospheric O_3 profiles with smaller spatial footprints has led to more research investigating the quality of the new UV-VIS satellites GOME-2 and OMI. First results indicate that indeed a considerable improvement has been made in terms of the accuracy of O_3 profile observations and the horizontal resolution of the measurements.

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7. *Was national or international cooperation established? If so, specify:*

Within TROPOSAT Dr de Laat has contributed to ACCENT-TROPOSAT-2, Task Group 1.
He is PI of the project "Tropospheric Ozone derived from Satellite measurements".
Within the TEMIS project requests have been received for tropospheric O_3 data from several international researchers, mainly for air quality and climate research. Contacts have been established with these researchers.

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8. *Have applications been found for the research results? If so, how can this be pursued?*

In this stage of the research to tropospheric ozone, applications are found in scientific research, in particular for the assimilation of O_3 profiles, which is not only relevant for air quality and atmospheric composition but also becoming more and more important for weather forecasting.
The TORA methodology will be applied to GOME-2 and OMI in the near future (TORA-2 proposal). Tropospheric ozone data products are relevant for user of satellite ozone data within the frameworks of O3SAF, GMES Atmosphere Service and the ESA project PROMOTE.

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9. *In case of doctoral research: when will the defence of the doctoral dissertation take place?*

N/A

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10. *What are the expectations with respect to the further career of the employee that was financed by SRON?*

The employee has performed excellent work within the TORA project, resulting in several publications and presentations at symposia. This has resulted in a growing network in the scientific community. The work has resulted in the start of new lines of research. One of the follow-up research plans has been submitted for funding to GO (TORA-2 proposal). Dr. de Laat will continue his research at KNMI later this year.

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11. *Publications:*
(incl. impact factor)

de Laat, A.T.J., R.J. van der A and M. van Weele, Evaluation of tropospheric ozone columns derived from assimilated GOME ozone profile observations, submitted to ACP, 2009. (ISI impact factor 4.86)

van Peet, J.C.A., R.J. van der A., A.T.J. de Laat and O. Tuinder, The height resolved ozone hole structure as seen by GOME-2, submitted to Geophys. Res. Lett., 2009. (ISI impact factor 2.602)

Laat, A.T.J. de, J. Landgraf, O. Hasekamp, I. Aben en B. Bregman, Validation of GOME ozone profiles and evaluation of stratospheric transport in a global chemistry-transport model. J. Geophys. Res., 2007, 112, doi:10.1029/2005JD006789. (ISI impact factor 2.8)

Mijling, B, O. Tuinder, R. van Oss, Preparing for GOME-2 ozone profileretrievals: improving the profile retrieval algorithm using GOME data, to be submitted to Atmospheric Measurement Techniques (AMT), 2009.

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12. *Lectures/congress papers/posters:*

Poster presentation:

van Weele, M, A.T.J. de Laat and R.J. van der A, Synergetic estimates of tropospheric ozone columns from total column observations and assimilated GOME profiles, Quadrennial Ozone Symposium (QOS), Tromso, Norway, June 29 – July 5, 2008.

van Weele, M, A.T.J. de Laat and R.J. van der A, Synergetic estimates of tropospheric ozone columns from total column observations and assimilated GOME profiles, IGAC 10th International conference, Annecy, Francem September 7-12, 2008.

Laat, A.T.J. de, M. van Weele, R. van der A, R. van Oss, O. Tuinder, B. Mijling, J. van Peet, H. Eskes en A. Segers, Determining tropospheric O3 columns using assimilated GOME O3 profiles Poster: Quadrennial Ozone Symposium 2008, 29/6/2008-5/7/2008, Tromso, Norway, International Ozone Commission (IOC) of IAMAS and the European Commission.

Presentations:

Laat, A.T.J. de et al., A new method to derive tropospheric ozone from GOME observationsPresentatie: 2006 EUMETSAT meteorological conference, 12/6/2006-16/6/2006, EUMETSAT (int.), Helsinki, Finland.

Laat, A.T.J. de, Role of tropospheric ozone increases in 20th-century climate change, KNMI KIK lunch, 8/6/2006, KNMI, de Bilt, The Netherlands.
Van der A, R.J, Data assimilation of Ozone from Satellite Observations, WMO O3 SAG, 27/6/2008, Tromsø.

De Laat, A.T.J. Tropospheric O3 column observations from satellites: what have we learned?, 18/9/2008, KNMI, de Bilt, The Netherlands.

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13. *Short summary (max. 500 words) of the initial aim and the tangible results* ***put into non-specialist, popular language*** *(this summary will be used for the ALW/SRON Annual Report, the PB-SRON website, and for reports to the funding ministries).*

Ozone is one of the most important atmospheric trace gases. About 90% of atmospheric ozone is located in the stratosphere between 10 en 50 km altitude, where it shields the lowest 10-15 km of the atmosphere from UV radiation which is harmful for life. The remaining 10% is located in the troposphere below 10 km altitude. Tropospheric ozone is for example required for the formation of the hydroxyl radical, also known as the atmospheric 'detergent'. Furthermore, tropospheric ozone is also one of the harmful atmospheric element in smog. Finally, tropospheric ozone is also an important greenhouse gas contributing about 20% to the current anthropogenic greenhouse gas radiative forcing. Scientists would love to continuously monitor the amount of tropospheric ozone. However, due to the scarcity of observational sites and the large variability of tropospheric ozone assessment of long-term tropospheric ozone trends and tropospheric ozone monitoring is difficult.

Satellite observations can fill this gap. However, because we are looking for 10% of the total amount of ozone BELOW the ozone layer where 90% of the ozone is located, it is difficult to 'see through' the ozone layer to determine the amount of ozone in the troposphere. It turns out that – in theory – satellites should be able to see just about the total amount of ozone in the troposphere, but only just, and no other errors may occur. Nevertheless, having this information already would contribute tremendously to our knowledge and monitoring ability of tropospheric ozone.

The key point in measuring tropospheric ozone from space is to separate the lowest 10 km of the atmosphere from the rest. Current direct observations of ozone can only achieve this more or less in the tropics. However, we would also like to learn more about tropospheric ozone outside of the tropics, for example in polluted industrialized countries in the Northern Hemisphere. Unfortunately this turns out to be difficult as explained and has been the one of the 'holy grales' of atmospheric remote sensing with satellites for more then 20 years now.

One method that has not been explored before is to combine many sources of information in a so-called assimilation procedure and produce the best estimate of the amount of ozone in the ozone layer. Since satellites can measure the total ozone column very accurately, subtracting the stratospheric column from the total column should leave a residual that would be an accurate estimate of the tropospheric ozone column.

This method was investigated in detail within the TORA project. The major finding was that indeed this methodology does work, but due to measurement and assimilation uncertainties and errors only in the tropics. Outside of the tropics the results are not useful – yet. Because the measurement and assimilation uncertainties can be – and to a certain extent in the last year have been - improved upon, we expect that the required accuracy can be achieved and that measuring tropospheric ozone columns comes well within reach. The results of the TORA project will be built upon during the coming years. The TORA-2 proposal is in the evaluation process.

Date: 26 February 2009

Signature Supervisor: Dr. Michiel van Weele

Signature Researcher(s):

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