Modelling Regional Differences in Tropospheric Ozone and Aerosols: The Reliability of Model Results

A contribution to subprojects GLOREAM and TOR-2

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Summary

The 3-D Eulerian grid model LOTOS (Long Term Ozone Simulation) that has originally been developed for hour-by-hour calculation of ozone formation over Europe, both for episodes and full year periods has been extended with the modules MADE (Ackermann et al. 2000) and SORGAM (Schell, 2000) for inorganic and organic secondary aerosols, respectively. The ozone-aerosol LOTOS-model should be considered as a model of intermediate complexity. The aim is to be able to perform many model runs over extended periods, but still keeping the major processes included in the model. The challenge in the LOTOS-model development and application is to decide and assess which simplification is still valid without loosing the reliability of the model and its results.

A data-assimilation technique has been developed and applied. The aim is to integrate observations and model results. In this way a more complete picture of the ozone and aerosol-fields over Europe can be established. Furthermore, weaknesses and inaccuracies in the model and its input can be detected and analysed, and the processes leading to regional differences over Europe can be studied in more detail.

Statistical methods have been developed and applied to investigate the reliability of the LOTOS-model results. Combining these methods with data-assimilation leads to an improved knowledge concerning model validation.

Introduction

The LOTOS-model has been extended and improved over the last years, with special attention to inorganic and organic aerosols and the incorporation of primary aerosol-emissions. Further improvements were focussed on boundary conditions, dry and wet deposition descriptions and land-use and land-cover, including biogenic emissions of VOC and NO. The aim still is to keep a balance between model simplification and reliability.

Data-assimilation for non-linear chemistry processes is a power-full tool to integrate observations and model results. It has been successfully applied in combination with LOTOS for both ozone and aerosols.

The statistical indices Fractional Bias (FB), Fractional Standard Deviation (FS), Normalised mean square errors of the Normalised Ratios (NNR) and several diagram methods have been used to analyse the behaviour of the LOTOS-model for ozone.

P.M. Midgley, M. Reuther (Eds.)
Proceedings from the EUROTRAC-2 Symposium 2002

Objectives

The aim of the research is to determine and analyse the processes which result in the differences in ozone and aerosol patterns over Europe over extended periods by using a model of intermediate complexity- including data-assimilation- and to determine the reliability of that model using also adequate statistical indices.

Activities and Results

Aerosol computations

Simulations with LOTOS for aerosols have been performed for the complete year of 1995 and for parts of 1997 and 2001. In general the model behaviour for aerosols seems quite adequate for SO_4: in most stations good correspondence with measurements is achieved. An example of a time series is given in Figure 1 for the station Kollumerwaard.

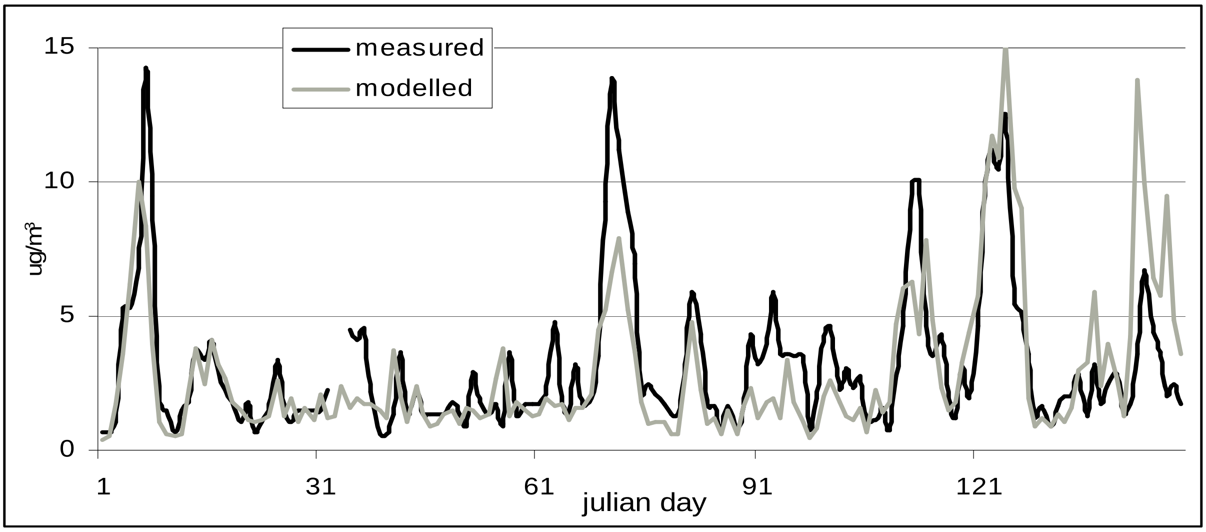

Figure 1. Measured and modelled (LOTOS) concentrations of SO_4 in the EMEP station NL09 (Kollumerwaard) for the first 150 days of 1995.

The same holds for NH_4 (ammonium). For nitrate the performance is less good, but still acceptable. The model seems not to be able yet to reproduce days with very high nitrate concentrations. When excluding these days the average modelled and observed nitrate levels are at approximately the same level. One must keep in mind that nitrate formation is a much more complex process. It strongly depends on the excess ammonia (i.e. the ammonia availabe after subtracting the double amount of sulfate on a molar basis) and on the temperature and relative humidity. Apart from ammonium-nitrate other forms of nitrate may be present that are not modelled yet.

Data assimilation

Data-assimilation has mainly been applied for a part of July/August 1997. Both aerosol and ozone concentrations have been assimilated. The aerosol measurements concerned SO4/NH4/NO3 and where taken from the EMEP network. The data assimilation technique is a special implementation of the Kalman Filter and the adaptions by the filter due to the measurements are based on the uncertainties in the model state. These uncertainties are the result of all kind of uncertainties in the input of the model, the model itself etc. As a modeller, one needs to identify and quantify these uncertainties and provide them to the data assimilation system. The system in turn is able to compute a ‘best fit’ based on the statistics of both the model and the measurements. In Figure 2 an example is given of the nitrate concentration at the Dutch station NL10, Vredepeel.

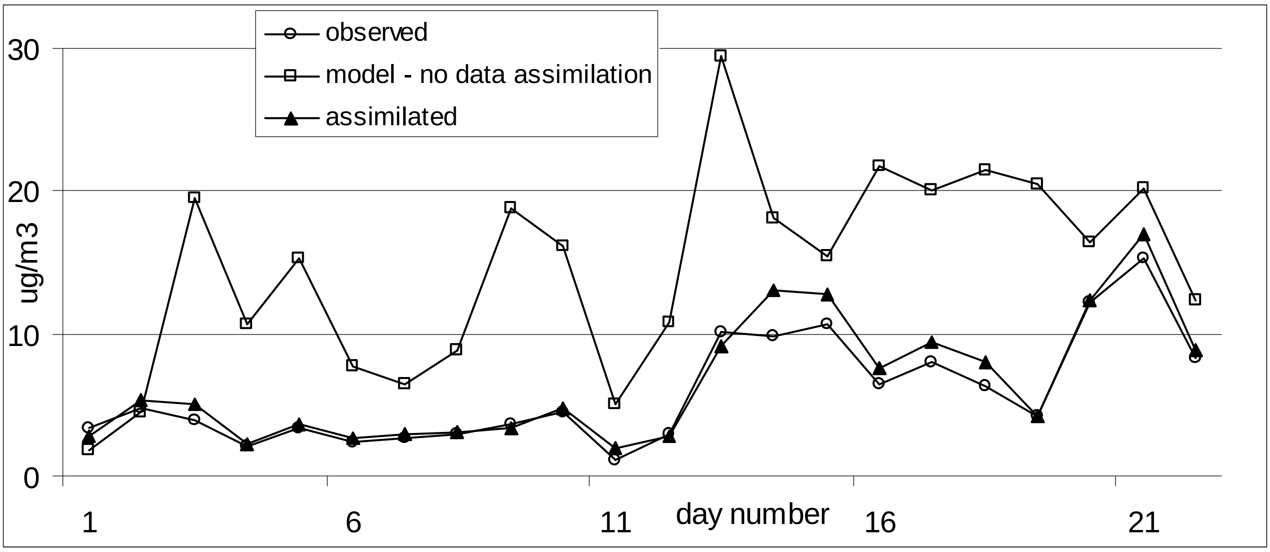

Figure 2. Measured and modelled concentration (with and without data assimilation) of nitrate (NO_3) for 28 July – 19 August 1997 at the EMEP station NL10 (Vredepeel)

From Figure 2 it can be seen that the modelled concentration of nitrate is too high in this period. However, with the specified model uncertainties – in this case in the emissions and the deposition velocities – the system is able to reach a good agreement between model and measurements. The change in the nominal emission input by the model is – on average – only minor, usually 5% or less. Only for NO_x adaptions are made up to 20%.

Statistical Analysis / Model validation

The statistical analysis has been applied to LOTOS-results for ozone for July and August 1995 using so-called “threshold” methods. The following indices have been used (the subscript *o* and *s* indicate “observed” and “simulated” respectively:

- the normalised accuracy of domain-wide maximum 1-hour concentration unpaired in space and time: $A_u = 100\left(\frac{C^s{}_{domain-wide\ peak} - C^o{}_{domain-wide\ peak}}{C^o{}_{domain-wide\ peak}}\right)$
- mean normalised bias of all simulated and observed concentration pairs with $C_i^o > 60\,\text{ppb}$: $NBIAS_{60} = \frac{100}{N}\sum_{i=1}^{N}\frac{\left(C_i^s - C_i^o\right)}{C_i^o}$

  where N includes all the simulated and observed concentration pairs with $C_i^o > 60\,\text{ppb}$
- mean normalised error of all simulated and observed concentration pairs with $C_i^o > 60\,\text{ppb}$: $NERROR_{60} = \frac{100}{N}\sum_{i=1}^{N}\frac{\left|C_i^s - C_i^o\right|}{C_i^o}$

Since A_u only makes sense for a relatively small domain, we have selected three areas for which the above norms have been calculated: 1. The south of the UK 2. the Benelux and northern Germany and 3. South Germany and a part of Switzerland.

The results show a good agreement between observations and model results in the Benelux and northern Germany, but an underestimation in the south of Germany and an overestimation in the south of the UK. Furthermore, the model tends to underestimate high concentrations

Conclusions

The current LOTOS model of intermediate complexity is a useful tool to investigate aerosol and ozone fields over Europe, both for episodic and long term calculations. Especially in the north-west of Europe the model performance is quite good for secondary inorganic aerosols and their precursors.

Data assimilation by Kalman Filtering has shown to be a powerful method for e.g. emission estimation. With relative minor adaptations to the emissions and deposition velocities the data assimilation system was able to improve the agreement between modelled and observed concentrations substantially. It seems that the selection of the stations used in the assimilation plays an important role. In order to see if there are biases in the model at certain locations or in certain areas, statistical analysis seems a good way to detect these in an objective way. Therefore the combination of data assimilation and statistical indices seems a promising method for model validation**.**

Acknowledgements

The TNO-part of the project is partly funded by the Dutch Min. of Economical Affairs via ECN. Drs J. Biswas, J.M. Davis, S.R. Hanna, and J.S. Irwin are grateful acknowledged to provide useful documents on statistical analysis. The contribution of E. Canepa was supported by the MURST COFIN 99 project ACME CUE, and by INFM PA project GEPAIGG01.

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