Can a relaxation technique be used to validate clouds and sulphur species in a GCM?

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SUMMARY

The Hamburg version of the European Centre for Medium-Range Weather Forecasting (ECMWF) general circulation model, ECHAM4, developed for climate studies, is used to study the hydrological cycle and the atmospheric sulphur cycle. A technique, the Newtonian relaxation, is introduced to compare results from the model with measurements of cloud parameters and sulphur species concentrations. The model is forced by ECMWF analyses to simulate September 1993, and results are compared to observations on different spatial and temporal scales: monthly mean horizontal satellite data, daily mean averages at European and Canadian stations and aircraft soundings over the North Atlantic off the Canadian coast. The model is able to reproduce monthly mean horizontal distributions of liquid water path and precipitation. Daily mean precipitation fluxes and the temporal evolution of gaseous and particulate sulphur also agree quite well with observations. Based on this comparison with observations the performance of two different cloud physics parametrizations is discussed. These experiments indicate that variables like relative humidity, cloud cover and precipitation are controlled by the large-scale dynamics, whereas the cloud water content depends on the parametrization of the cloud microphysics. Coupling of the model-calculated sulphate mixing ratios to the cloud microphysics improves the model’s sulphate distribution but slightly worsens the agreement between calculated and observed precipitation.

KEYWORDS: Hydrological cycle Model evaluation Observations Sulphur cycle

1. INTRODUCTION

General circulation models (GCMs) used for climate studies have to be integrated over long periods, typically 30 years, to obtain statistically significant averages. Such models are evaluated by comparing multi-year averages and the models interannual variability to large-scale synoptic observations. It is difficult to evaluate model parameters, like cloud cover or cloud liquid water, when most of the data are short-term observations. Clouds have an important impact on the climate system: directly by changing the radiative fluxes, and indirectly by acting as chemical reactors and by modifying the atmospheric aerosol concentration. Cloud feedback, which differs considerably between current models (Cess et al. 1996), is a key issue for climate dynamics, so there is a real need to carefully evaluate the parameters of the hydrological cycle. Moreover, due to an increasing interest in the possible impact of climate of enhanced emissions of aerosol precursor substances in recent years, chemistry schemes have been implemented in GCMs, and concentrations of chemical species have also to be evaluated.

We performed simulations of the atmospheric sulphur cycle in the Hamburg climate model ECHAM (ECMWF model HAMburg version) to study feedback mechanisms between climate and atmospheric chemistry (Feichter et al. 1996; Feichter et al. 1997, Lohmann and Feichter 1997). As the database of chemical species is much smaller than for any meteorological parameter, models can be validated only over some specific regions like Europe or North America, where surface data are available. Vertical profiles of chemical species can only be compared with observations taken during measurement campaigns. Additionally, since most of the chemical species of interest have atmospheric residence times of the order of days, we have to carefully check the model’s ability to produce reliable results on this time-scale. Even a weather forecast model is
not the appropriate tool to simulate the chemical species concentrations associated with a specific weather episode, because no data assimilation technique, as used in numerical weather prediction, is available to initialize the chemistry model. Therefore, we apply a technique, Newtonian relaxation (or ‘nudging’), to force the model to simulate a specific weather episode. This technique allows us to integrate the model over such long time intervals that the initial conditions do not affect the chemical species distributions, and it enables model validation by comparing simulated with observational data. Transport processes of the short-lived tracer Radon-222 have been studied in a companion paper by Dentener et al. (1999). This paper focuses on the comparison of cloud and sulphur species from three simulations with the ECHAM GCM. The first one is obtained from an experiment which is forced only with the observed sea surface temperature (SST). In the second experiment the nudging technique is applied. In both experiments the sulphur cycle cannot feedback on clouds and radiative-transfer schemes. As cloud formation and lifetime is strongly influenced by the abundance of aerosols, because aerosol particles act as cloud condensation nuclei (CCN), another nudging experiment is performed. In this experiment the mass of sulphate aerosols determines the number of cloud droplets and, hence, can change the lifetime of clouds. Thus, as the cloud lifetime is crucial for sulphate formation (80% of sulphate is formed in cloud droplets; Feichter et al. 1997), and the atmospheric residence time of sulphate particles is mainly determined by removal by rain and snow, a new feedback loop is introduced. Thus, the question is, whether an explicit consideration of the impact of aerosols on the cloud physics results in a better representation of the hydrological cycle, because there is a strong coupling between cloud processes and sulphur chemistry.

The climate model, the sulphur chemistry and the cloud physics are described in section 2. In section 3 zonal mean distributions of liquid water path and precipitation as calculated by the assimilated and non-assimilated ECHAM experiments are compared to observations from satellite microwave measurements (Special Sensor Microwave Imager, SSM/I) and the global precipitation climatology project (GPCP). In section 4 we compare surface measurements of precipitation fluxes and sulphur concentrations to observations reported by EMEP (the European Monitoring and Evaluation Programme; Hjelbrekke et al. 1995) and to sulphur measurements from CAPMON (the Canadian Air and Precipitation Monitoring Network). Both networks report daily mean mixing ratios and the sites of CAPMON are located in the same latitudes as the EMEP network. Section 5 focuses on the comparison of simulated vertical profiles with aircraft observations obtained during the North Atlantic Regional Experiment (NARE) in September 1993.

2. Model Description

The model used in this study is the GCM ECHAM4 in T42 (truncation at wavenumber 42) horizontal resolution (approximately 2.8° × 2.8°). Discretization in the vertical is done by using a hybrid coordinate system with 19 levels, with its uppermost level at about 30 km. The boundary layer is resolved by five levels corresponding with heights of ~ 30, 150, 390, 800, and 1400 m. The observed daily mean SSTs are used as lower boundary conditions. A general description of the model can be found in Roeckner et al. (1996). We applied the Newtonian relaxation method (Krishnamurti et al. 1991; Jeuken et al. 1996) to adjust ECHAM to observations. This technique relaxes the model state toward observational data by adding, at each time step, an additional term to the model’s
equations:
\[ \frac{\partial X}{\partial t} = TE_X + G_X \cdot (X_{\text{obs}} - X). \]

Here \( X \) represents a model-calculated prognostic variable, \( TE \) is the tendency of \( X \) as calculated by the model's equation, \( G(s^{-1}) \) is the relaxation coefficient which defines a characteristic adjustment time-scale, \( X_{\text{obs}} \) is the observed value of \( X \) and \( t \) is time. In this experiment we relaxed the wind (vorticity and divergence), the surface pressure and the temperature to ECMWF analyses. Jeuken et al. (1996) performed experiments with a coarser resolution version of ECHAM (ECHAM3-T21) and recommended the following relaxation coefficients which are also used in the current study:
\[ G(\text{Vorticity}) = 1.10^{-4} \text{ s}^{-1}, \quad G(\text{Divergence}) = 0.5 \cdot 10^{-4} \text{ s}^{-1}, \quad G(\text{Temperature}) = 1.10^{-5} \text{ s}^{-1} \]
and
\[ G(\text{Surface pressure}) = 1.10^{-4} \text{ s}^{-1}. \]

The cloud microphysics scheme implemented in the operational version of ECHAM (OPER) is described in detail in Roeckner et al. (1996), and the scheme which couples aerosol distribution with the cloud physics (COUP) in Lohmann and Roeckner (1996) and Lohmann and Feichter (1997). In OPER, cloud water and cloud ice are treated as a common prognostic variable, whereas in COUP they are treated separately. In COUP the bulk microphysics parametrizations for warm-phase processes are derived from the stochastic collection equation (Beheng 1994), while the parametrizations of the mixed and ice phase were originally developed for a mesoscale model (Levkov et al. 1992). Parametrized microphysical processes are: condensational growth of cloud droplets; depositional growth of ice crystals; homogeneous and heterogeneous freezing of cloud droplets; autoconversion of cloud droplets; aggregation of ice crystals; accretion of cloud ice and cloud droplets by snow; accretion of cloud droplets by rain; evaporation of cloud water and rain; sublimation of cloud ice and snow; and melting of cloud ice and snow. The advection of cloud water and cloud ice is omitted. A parametrization of the autoconversion rate of cloud droplets (Beheng 1994), which depends not only on the liquid water content as in OPER but additionally on the cloud droplet number concentration (CDNC), is introduced. At present CDNC cannot be computed realistically in GCMs because it depends on several factors which are not easy to predict, such as subgrid-scale vertical velocity, maximum supersaturation, and availability of CCN. Therefore we relate CDNC empirically to the sulphate aerosol mass (Boucher and Lohmann 1995). Measurements of sulphate (SO$_4^{2-}$), CCN and CDNC have been taken at various continental and marine sites in clean and polluted air, for a variety of weather situations. Fractional cloud cover is an empirical function of the relative humidity (Sundqvist 1989).

The atmospheric sulphur cycle is simulated on-line with the meteorology; it treats emission, transport, dry and wet deposition and chemical transformations of sulphur species. Prognostic variables are dimethyl sulphide (DMS) and sulphur dioxide (SO$_2$) as gases and SO$_4^{2-}$ as an aerosol. Biogenic emissions from the oceans and from soils and plants are assumed to occur as DMS (19 Tg sulphur per year), while emissions from volcanoes, from biomass burning, and from combustion of fossil fuel and from smelting occur as SO$_2$ (77.3 Tg sulphur per year). DMS, as well as SO$_2$ in the gaseous phase, is oxidized by reaction with the hydroxyl radical (OH) during the day. Additionally, DMS reacts with nitrate radicals (NO$_3^-$) at night. We have assumed that the only end product of DMS oxidation is SO$_2$. Dissolution of SO$_2$ within cloud water is calculated according to Henry's law. In the aqueous phase we consider oxidation of SO$_2$ by hydrogen peroxide (H$_2$O$_2$) and ozone (O$_3$). Three-dimensional monthly mean oxidant concentrations are prescribed based on calculations using ECHAM and a more comprehensive chemical
model (Roelofs and Lelieveld 1995). The calculation of the reaction rate with O$_3$ and the effective Henry's Law constant for SO$_2$ requires assumptions about the cloud pH. Assuming that aqueous phase equilibria and ion balance are maintained, [H$^+$] is approximated by assuming a molar ratio of 1 between sulphate and ammonium (Dentener and Crutzen 1994), where the square brackets indicate molecules cm$^{-3}$.

The following three simulations have been performed:

- AMIP (Atmospheric Model Intercomparison Project): Operational ECHAM4, non-assimilated.
- OPER: Operational ECHAM4, assimilated, sulphur species do not influence any meteorological parameter.
- COUP: ECHAM4 with new cloud microphysics (Lohmann and Roeckner 1996), assimilated, sulphate aerosol mass determines the number of cloud droplets.

All experiments have been performed with prescribed SST and have been integrated from January to September 1993.

3. **Comparison with global distributions of meteorological variables**

Quantities such as wind, temperature and surface pressure which have been used to assimilate the model are in good agreement with the ECMWF analyses, as expected. Therefore, we concentrate the discussion on parameters which have not been adjusted to the ECMWF analyses, like cloud liquid water and precipitation. In the following, global distributions of monthly mean averages representative of the month of September 1993 will be discussed. Precipitation observations are taken from the GPCP (version 1) (Huffman et al. 1997). This time- and area-averaged precipitation dataset is based on all suitable observations, using mainly rain-gauge measurements over the continents and satellite retrievals over the oceans. The SSM/I observations of liquid water path (LWP) and the precipitation fluxes are highly uncertain (Chen et al. 1996).

Figure 1 presents the zonal mean distribution of LWP for September 1993 from OPER and COUP, and for SSM/I data retrieved by Weng and Grody (1994). Additionally SSM/I data from Greenwald et al. (1993), which represent a five-year ensemble mean of September data, are shown to illustrate the uncertainty associated with these SSM/I data. The SSM/I data from Weng and Grody (1994) are 60% lower in the global mean (53 g m$^{-2}$) than the SSM/I data from Greenwald et al. (1993) (79 g m$^{-2}$), so that the comparison with the observations can only be done qualitatively. The LWP comparison is restricted to oceans between 60°N and 60°S, because LWP cannot be retrieved over land and ice-covered ocean due to the large variation in surface emissivity. The observed LWP shows maximum values in the intertropical convergence zone (ITCZ) at 10°N (between 70 g m$^{-2}$ for Weng and Grody and 90 g m$^{-2}$ for Greenwald et al.) and up to 110 g m$^{-2}$ associated with cyclonic activity over the northern hemisphere (NH) midlatitude oceans. LWP in the southern hemisphere (SH) midlatitudes is smaller than over NH with maximum values up to 90 g m$^{-2}$. The dry subtropical subsidence regions are clearly marked in the SSM/I retrieval from Weng and Grody, but less pronounced in Greenwald et al. To compare these observations with OPER and AMIP we derived LWP from the total cloud water mixing ratio as a function of the temperature (Rockel et al. 1991). COUP calculates cloud liquid water mixing ratio as a prognostic variable. In all simulations the global mean LWP over the oceans is between the SSM/I retrieval from Weng and Grody, representing September 1993, but lower than the retrieval from Greenwald et al. which represents a 5-year September average. The monthly and global mean
is lowest in OPER (59 g m$^{-2}$), amounts to 67 g m$^{-2}$ in AMIP and is highest in COUP (77 g m$^{-2}$). The simulations (Fig. 1) capture the main features of the observations. However, in the NH midlatitudes COUP overpredicts LWP. This overprediction is due to the high sulphate particle concentrations downstream of the polluted industrial regions of the eastern USA and south-east Asia, and hence the high concentration of cloud droplets. Thus, the autoconversion of cloud droplets to form rain drops is slower and, therefore, the lifetime of those clouds prolonged (Lohmann and Feichter 1997). Furthermore, all model versions simulate the LWP in the ITCZ at 10$^\circ$N and indicate a second maximum in the tropics at 10$^\circ$S which is not shown in the observations. LWP is overestimated over the subtropical oceans of the NH with respect to the SSM/I observations of Weng and Grody in all simulations. In COUP the predicted LWP is also larger than that suggested by Greenwald et al. OPER and AMIP fall in between both retrievals. This emphasizes that the accuracy of current satellite retrievals is not sufficient to evaluate the simulated LWP.

A comparison of the GPCP precipitation data with the simulations for September 1993 is shown in Fig. 2. Maxima in the observed precipitation are located in the ITCZ and over the storm tracks in midlatitudes. Large differences exist between the assimilated and non-assimilated experiments, whereas only small differences are apparent between the two assimilated experiments. In all simulations the equatorial belt of high precipitation is too strong and too narrow and is shifted northward by about 5$^\circ$. Overall the precipitation simulated in the AMIP experiment agrees best with the observations. Its ITCZ is not as narrow and the amount of precipitation in midlatitudes is larger than in the assimilated experiments. In the global mean the precipitation amounts to 2.8 mm day$^{-1}$ in AMIP, 2.7 mm day$^{-1}$ in OPER and 2.6 mm day$^{-1}$ in COUP compared to 2.7 mm day$^{-1}$ in the GPCP data.

In summary: the comparison of zonal mean precipitation and LWP for September 1993 does not reveal any superiority of the assimilated over the non-assimilated experiments. The monthly mean precipitation and, therefore, the hydrological cycle of both
assimilated ECHAM versions (OPER and COUP) is weaker than the hydrological cycle of the non-assimilated version (AMIP). Thus, the location and amount of precipitation is strongly governed by dynamical processes, and the microphysical processes are only second-order effects. LWP, however, in both assimilated versions deviates in opposite directions from AMIP, as it is determined by the microphysical processes rather than dynamical processes. Jeucken et al. (1996) have shown, based on tracer studies, that the vertical exchange between the boundary layer and the free troposphere is lower if ECHAM is assimilated. This may also slightly weaken the vertical exchange of latent heat, and damp the hydrological cycle.

4. COMPARISON WITH SURFACE MEASUREMENTS

For September 1993 the EMEP database (Hjelbrekke et al. 1995) provides daily mean measurements of precipitation and the concentration of sulphur dioxide and sulphate at 190 sites in Europe. We used these data to evaluate the predictive skill of the assimilated model versions and their ability to simulate the spatial and temporal evolution of weather systems. September 1993 is characterized by relatively high pressure over Greenland and Iceland and by low pressure over western Europe. The frontal zone is extended from Spain over the southern edge of the Alps to the Black Sea. Over Central Europe we find a weak high-pressure system. Negative temperature anomalies occur over northern Europe and positive anomalies over south-east Europe. During the first half of September heavy precipitation occurred over Germany and the southern Alps and during the second half over northern Italy and southern France.

Figure 3 shows the frequency distribution for different precipitation classes at all observation sites. Days without precipitation are observed twice as often as predicted by the model, whereas precipitation in the range up to 15 mm day\(^{-1}\) is overpredicted by the model. This may be due to the fact that we are comparing model results representing an average over an area of about 150 \(\times\) 150 km\(^2\) with point measurements. The occurrence of precipitation over this relatively large area is of course more likely than over a specific
site. Additionally, the measurements are not representative of such a large area. It has been estimated that (WMO, 1985) 12 stations are necessary to obtain total monthly precipitation with an accuracy of 10% on a 2.5° × 2.5° grid. The uncertainties may be higher over mountainous terrain. However, the accumulated precipitation of all classes up to 15 mm day\(^{-1}\) agrees perfectly with the EMEP data. A reasonable agreement between predicted area averages and the EMEP observations is found in the precipitation classes between 15 and 30 mm day\(^{-1}\). The bias increases again for daily precipitation amounts higher than 30 mm. In this range the model underestimates the precipitation, and no precipitation events higher than 40 mm day\(^{-1}\) are simulated by the model. Such high precipitation amounts would not be realistic as an area average, either. It is interesting to note that the frequency distribution of precipitation is not altered by nudging or by a different cloud microphysics scheme.

The skill of the model versions in simulating the precipitation correctly is presented in Table 1. We count at every site the number of events where the observed daily precipitation is less than or equal to 0.1 mm day\(^{-1}\) or more than 0.1, 1.0, 2.5 and 10 mm day\(^{-1}\). Additionally, the percentage of correctly predicted amounts of daily precipitation is calculated. Daily precipitation amounts less than 0.1 mm are only captured by the model in less than 50% of the cases, whereas precipitation events in the three ranges larger than 0.1 mm day\(^{-1}\) are simulated well by the model in 79%, 76% and 69% of the cases, respectively. The percentage of scores decreases for stronger events, and the rate of correct predictions is at most 15% for precipitation events greater than 10 mm day\(^{-1}\). The precipitation characteristics are clearly improved when the model is assimilated.

The bias between the model calculations and the EMEP observations of the daily precipitation amount averaged over the month for every measurement site is presented in Fig. 4. The relative bias used in Fig. 4 is defined as \((m - o)/(m + o)\) with \(o\) denoting the mean daily observations and \(m\) the model results. Values between \(-0.33\) and \(+0.33\) denote factor-of-two differences between model and observation. Figure 4 shows that the model overpredicts the precipitation at about 85% of the sites, although the observed
TABLE 1. PERCENTAGE OF CORRECTLY PREDICTED DAILY PRECIPITATION AMOUNTS

<table>
<thead>
<tr>
<th>Precipitation classes (mm day⁻¹)</th>
<th>Number of events</th>
<th>Model predictions: Scores (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>EMEP</td>
<td>OPER</td>
</tr>
<tr>
<td>no observations available</td>
<td></td>
<td></td>
</tr>
<tr>
<td>≤0.1</td>
<td>122</td>
<td>47</td>
</tr>
<tr>
<td>&gt;0.1</td>
<td>912</td>
<td>79</td>
</tr>
<tr>
<td>&gt;1.0</td>
<td>710</td>
<td>62</td>
</tr>
<tr>
<td>&gt;2.5</td>
<td>548</td>
<td>49</td>
</tr>
<tr>
<td>&gt;10</td>
<td>238</td>
<td>15</td>
</tr>
</tbody>
</table>

Figure 4. Relative bias between observed (EMEP) and calculated precipitation defined as \((\text{obs-model})/(\text{obs+model})\) plotted against height; for details of models OPER, COUP and AMIP (see text).

The mean precipitation amount averaged over all the sites is higher (3.5 mm day⁻¹) than the calculated one (OPER: 2.0; COUP: 2.2; AMIP: 2.3 mm day⁻¹). This is in agreement with the frequency distribution in Fig. 3, which shows an underprediction of strong precipitation events and an overprediction of weak precipitation. The assimilated model versions, OPER and COUP, predict the precipitation within a factor of two at 86% and 82% of all sites, respectively, whereas in AMIP, where only the SST is prescribed, 61% are within a factor of two of the observations. The bias in Fig. 4 is plotted against height above sea level and shows a smaller bias at sites above 500 m. This is probably due to the fact that observed precipitation amounts increase with height due to orographic effects whereas calculated amounts do not, because the model does not resolve such small-scale orographic effects. As the model overestimates the precipitation amount, the agreement seems to be better at higher altitudes. No dependency of the bias on latitude or longitude, i.e. on the geographical location, is found.

In Fig. 5 the temporal correlation between the observed and the calculated daily means at every site is shown. Note that only the 67 sites which have weather reports throughout the month and have reported or simulated at least one precipitation event are considered. Here we see a better correlation at sites at lower altitudes since the
model cannot resolve orographic effects. The temporal correlation shows a distinct
difference between the assimilated and the climatological ECHAM versions, with an
average correlation coefficient $r = 0.37, 0.33$ and $0.17$ for experiments OPER, COUP
and AMIP, respectively. The spatial correlations calculated for every day and averaged
over the month are slightly worse, $r = 0.26, 0.24$ and $0.13$, respectively. Nevertheless,
these correlation coefficients are within the range of that between observations within
one grid box, because precipitation is highly variable both in time and space.

The agreement may be better if we run the model with finer horizontal resolution.
Stendel and Arpe (1997) compared results of the T106 version of the ECMWF weather
forecast model at one grid box in central Belgium, covered by 31 observing stations,
and found that July daily precipitation amounts over a seven year period produced a
correlation coefficient of 0.7 between the model and the observational area average.
However, the station density of the EMEP data is not good enough to analyse the
precipitation on a regular grid, so we are limited to comparisons at specific sites.

The comparison of daily mean surface-air concentrations of SO$_2$ and SO$_4^{2-}$ between
observations and model simulations is a rather difficult task for a relatively coarse-
resolution global model. Concentrations of short-lived species like SO$_2$ or SO$_4^{2-}$ cover a
wide range of two orders of magnitude. We present in Table 2 the bias and the correlation
coefficients to compare the mean daily observations with the model results; additionally
the ‘Spearman rank correlation’ is chosen as a robust test resistant to outlying data points
to determine whether correlations between the observations and the model results are
significant.

The calculated SO$_2$ concentrations are higher by a factor of two than the EMEP
data at all 34 sites; SO$_4^{2-}$ is predicted within a factor of two at 76% and 38% of the
stations (42 sites) by OPER and COUP, respectively. The model behaves similarly over
Canada, although the agreement with CAPMON observations is slightly better than
with the EMEP data. At the 10 sites reported by CAPMON the model calculates SO$_2$
mixing ratios only at two sites within a factor of two of the observations, whereas the
calculated SO$_4^{2-}$ is at all sites within a factor of two, with a tendency to underestimate
TABLE 2. COMPARISONS OVER EMEP AND CAPMON AREAS OF OBSERVED SO₂ AND SO₄²⁻ WITH MODEL RESULTS

<table>
<thead>
<tr>
<th>Species</th>
<th>Models</th>
<th>EMEP</th>
<th>CAPMON</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Relative Bias</td>
<td>r</td>
</tr>
<tr>
<td>SO₂</td>
<td>OPER</td>
<td>0.79</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td>COUP</td>
<td>0.59</td>
<td>0.43</td>
</tr>
<tr>
<td>SO₄²⁻</td>
<td>OPER</td>
<td>0.14</td>
<td>0.22</td>
</tr>
<tr>
<td></td>
<td>COUP</td>
<td>0.40</td>
<td>0.42</td>
</tr>
</tbody>
</table>

Relative bias is averaged over the measurement sites and the daily means; correlation coefficient (r) is between observed and calculated daily means averaged over the measurement sites; number of measurement sites in % whose time series (30-day means) are correlated at the 95% confidence level (c.l.) using the Spearman rank correlation coefficient, significantly negative correlations are given in parenthesis.

The observations. As discussed in Feichter et al. (1996) the chemistry model tends to overpredict the SO₂ surface concentrations, in particular over high latitudes, maybe due to too slow an oxidation rate or due to weak vertical exchange. Additionally, the anthropogenic sulphur emissions used in our simulations are representative for the year 1985, but sulphur emissions in Europe declined strongly during later years. Berge et al. (1995) report a decrease in sulphur emissions over Europe by about 30% from 1985 to 1993. In Germany, where 22 of the 34 EMEP stations (reporting SO₂ concentrations) are located, the emissions of SO₂ decreased from 7754 to 3896 kt year⁻¹. Furthermore, the emissions in highly populated regions show a strong temporal and spatial variability, whereas our source distribution represents a seasonal mean with a coarse spatial resolution. Consequently, the agreement between model and observation is worse in central Europe. Anthropogenic emissions over Canada have not changed significantly between 1985 and 1993.

The agreement is somewhat better if we compare the temporal correlation between the daily mean concentrations, indicating that the day-to-day variability at a specific site is captured better than the magnitude of the concentrations. In Fig. 6 the correlation coefficients of surface SO₂ and SO₄²⁻ over Europe are plotted against the longitude. Both species concentrations show a better agreement with EMEP in the COUP model version compared to OPER. The mean correlation for SO₂ is 0.40 and 0.43 for OPER and COUP, respectively, and the corresponding correlation coefficients for SO₄²⁻ are 0.22 and 0.42. Whereas the correlation of SO₂ does not show a longitudinal dependency, SO₄²⁻ as calculated by OPER performs distinctly worse between 7⁰E and 20⁰E. Over Canada the correlation of SO₂ between observations and model is 0.33 and 0.43 for OPER and COUP, respectively, which is worse than over Europe, whilst that of SO₄²⁻ is 0.42 and 0.44 and better than over Europe.

The Spearman rank correlations between the 30-day observed and calculated time series of SO₂ mixing ratios are significant at the 95% level at 49% and 46% of the 34 EMEP sites and at 40% and 60% of the 10 CAPMON sites for OPER and COUP, respectively. The observed and calculated time series of SO₄²⁻ are significantly correlated at 40% and 79% of the EMEP sites and at 50% and 60% of the CAPMON sites for OPER and COUP, respectively.

The comparison with daily mean surface observations suggests a relatively good agreement between calculated and observed precipitation fluxes. The superiority of the
assimilated experiments is clearly seen in much higher scores of the amount of daily mean precipitation. The new cloud microphysics scheme (COUP), however, performs slightly worse compared with the operational scheme (OPER).

The temporal correlation between simulated and observed sulphur species is satisfactory, however the actual concentrations are not. In particular, the SO$_2$ concentrations are much too high in the model simulations, and SO$_4^{2-}$ is also overestimated. However, for a realistic simulation of chemical cycles on such small temporal and spatial scales, a higher resolution of the emission database is mandatory. The coupled cloud physics–sulphur scheme (COUP) shows a better agreement with the observed sulphur concentrations, in particular the sulphate mixing ratios over Europe are significantly improved compared to OPER.
5. COMPARISON OF VERTICAL PROFILES

To investigate the performance of the model in simulating the vertical distributions of clouds and tracers in and above the planetary boundary layer, we compare our model simulations to vertical profiles taken from the August–September 1993 measurement campaign 'North Atlantic Regional Experiment' (NARE). Clouds were encountered on only six days during the campaign, from 2 to 8 September (Banic et al. 1996). Since the focus of our study is on cloud–aerosol interactions, we have chosen this period to compare the observational data to our model results. Leaitch et al. (1996) conducted six flights from 4 to 7 September within a 100 km radius of Yarmouth (43.5°N, 66.1°W), Nova Scotia, Canada. Profiles of temperature, humidity, cloud liquid water content, cloud droplet and aerosol particle number concentrations and SO$_2$ have been reported.

Patchy fog and low stratus developed during the afternoon of September 5 and covered the sampling area. Stratus below 800 m formed as a result of the cooling of warmer and moister air originating further south. These conditions persisted throughout 5 to 7 September. According to trajectory analyses (Liu et al. 1996), the air sampled on 6 September spent some time over the Nova Scotia peninsula, whereas the air on 7 September had previously been over the sampling area on 4 September.

The results presented here are only from the two assimilated model simulations. Vertical profiles taken at Yarmouth are shown in Fig. 7 and compared to the model predictions. We present soundings from 1830 UTC 7 September. During this measurement flight all parameters of interest have been reported, and the observational time coincides with the model output time. Wind, temperature and relative-humidity profiles of both experiments agree quite well with the aircraft measurements, except close to the surface. The model is not able to simulate an inversion layer at about 500 m altitude, but instead calculates a linear temperature decrease with height (Fig. 7(a)). The increase of wind speed with height is captured, but the wind in the boundary layer is too strong (Fig. 7(b)), which may explain why no temperature inversion forms in the model (Fig. 7(c)). The relative-humidity profiles are too dry above 800 hPa. The profiles in OPER and COUP suggest a lower relative humidity everywhere. While OPER simulates a higher relative humidity in the boundary layer than COUP, which is in better agreement with the observations, it is much dryer above 900 hPa. Thus, the vertical gradient in relative humidity is too large in OPER. Below the temperature inversion a thin stratocumulus layer and a thin layer of fog were observed. Both in OPER and COUP cloud water is lower than that observed by more than one order of magnitude (Fig. 7(d)). In contrast to the cloud water profiles, the simulated cloud cover, which is calculated diagnostically based on the relative humidity (Sundqvist 1978), shows a maximum at 400 m, in better agreement with the observations (Fig. 7(e)). COUP predicts a peak in cloud droplet number concentrations at the correct altitude, but overestimates the number by about 50% at that particular time (Fig. 7(f)). The observed SO$_2$ profile shows a maximum of 4 parts per billion by volume (p.p.b.v.) at 870 hPa and of 2 p.p.b.v. at 600 hPa (Fig. 7(g)). OPER and COUP predict high mixing ratios within the planetary boundary layer (PBL) in contrast to the observations, although they do predict a linear decrease above PBL in agreement with the observations. The horizontal distributions of the model’s SO$_2$ exhibit high mixing ratios over Pennsylvania, and a tongue of high sulphur concentration extends along the east coast of the USA and Canada. Thus the horizontal gradients are strong over the Yarmouth region resulting in large differences between neighbouring grid points. SO$_4^{2-}$ profiles are presented in terms of aerosol particle numbers assuming a log-normal size distribution with a mean radius of 0.07 µm, a standard deviation (σ) of 2.03 and a density of 1.7 g cm$^{-3}$ for dry SO$_4^{2-}$. These number concentrations are compared with
Figure 7. Calculated and observed vertical profiles at 1800 UTC 7 September 1993, at Yarmouth of: (a) temperature (K), (b) wind (m s\(^{-1}\)), (c) relative humidity (%), (d) cloud liquid water content (g m\(^{-3}\)), (e) cloud cover (%), (f) number of cloud droplets (cm\(^{-3}\)), (g) SO\(_2\) mixing ratio (p.p.b.v.), (h) and total number of aerosols (cm\(^{-3}\)). Observed profiles are from the North Atlantic Regional Experiment (NARE; full lines) calculated profiles are from model simulations OPER (dotted lines) and COUP (dashed lines). See text for further details.
measurements performed with a passive cavity aerosol spectrometer probe (PCASP) which measures particles in the size range between 0.135 to 3.0 μm (Liu et al. 1996). Leaitch et al. (1996) found a rather high maximum particle number concentration above the stratocumulus cloud of 3800 particles cm$^{-3}$ indicating anthropogenic polluted air masses from the North American continent. Very clean air with particle concentrations lower than 200 N cm$^{-3}$ was observed below 400 m and above 3 km altitude (Fig. 7(h)). The layer with high particle concentration showed a sharp decrease in concentration below 930 hPa which is about the same height as the top of the cloud. This may be due to the fact that particles taken up by cloud droplets are not counted by the PCASP detector (Kleinman et al. 1996). Both model experiments predict relatively low particle concentrations of 300 to 370 N cm$^{-3}$ within the lowest km and show a linear decrease in the upper levels. Even if we take into account that, according to Liu et al. (1996), during the NARE campaign only 43% of the total aerosol mass in the accumulation mode is SO$_4^{2-}$, the model badly underestimates the particle concentrations.

To summarize: the comparison of model simulations with aircraft measurements yields acceptable agreements of wind, temperature and relative humidity, but the agreement is worse for cloud water and sulphur concentrations which exhibit small-scale structures and consequently a high temporal and spatial variability. In particular, due to its coarse vertical resolution the model cannot resolve the thin inversion layer near the surface which is, according to the observations, decoupled from the layers above.

6. DISCUSSION AND CONCLUSIONS

Newtonian relaxation has been applied in the climate model ECHAM4 to simulate a specific meteorological episode, the month of September 1993. For this purpose, surface pressure and the three-dimensional wind and temperature fields were adjusted to ECMWF analyses. This method allows the evaluation of climate-model performance on smaller temporal scales and comparison with short-term observational data. Since ECHAM4 will be used to calculate short-lived trace gas and aerosol particle distributions it is particularly important that the model behaves well, not only on climatological time-scales—defined as 30-year averages—but also on time-scales on the order of days. Hence, the aim of this study was to demonstrate the ability of the model to reproduce the hydrological cycle and the distribution of some sulphur species on different spatial and temporal scales.

Three experiments have been performed: one with prescribed monthly mean SST but no further adjustment (AMIP) and two experiments with relaxation of wind, surface pressure and temperature to observations, one with the operational cloud physics (OPER) and one with new cloud microphysics which relates the calculated sulphate mass to the number of cloud droplets (COUP). Results from these models have been compared to global monthly mean horizontal distributions derived from satellite data and ground based observations, to daily averages taken from the EMEP and the CAPMON databases, and to vertical profiles taken from aircraft measurements performed during NARE.

First, we compared horizontal monthly mean distributions of liquid water path and precipitation with observations. The liquid water path of the assimilated and of the non-assimilated model versions looked very similar. This may be due to the fact that even in the non-assimilated version the observed SST is prescribed and satellite data are only retrieved over the sea. The liquid water paths are mostly between the two observational estimates. Precipitation fluxes agreed very well between observations and all model
versions except for the ITCZ where all model versions shift the maximum about 5° further north.

Comparing the three model versions we find that the distribution of precipitation flux of the assimilated versions (OPER and COUP) looks very similar. The same holds for relative humidity and total cloud cover. The assimilation method weakens the subgrid-scale vertical exchange as shown by Jeukcn et al. (1996) and dampens slightly the hydrological cycle, resulting in lower precipitation. The liquid water path in COUP is significantly higher than in OPER due to aerosol–cloud interactions. This indicates that variables like relative humidity, cloud cover (which is parametrized depending only on relative humidity) and precipitation flux are mainly determined by the large-scale dynamics, whereas the cloud water content depends crucially on the choice of the cloud microphysics parametrization.

Next, we compared simulated precipitation fluxes, SO_2 and SO_4^{2-} concentrations to daily mean averages over Europe and Canada. The agreement is good, considering that a model using such a coarse grid cannot reproduce extreme events. Events with no precipitation over a day are less likely to be simulated within a model grid-box than they are to be observed at a specific measurement site. Analysis of the daily precipitation fluxes at 190 sites showed that 86% and 82% of the sites using assimilated model versions OPER and COUP, respectively, are within a factor of two of the observations, compared with only 62% of all sites in the non-assimilated version, AMIP. The temporal correlation between observations and calculations averaged over all the 67 sites yields correlation coefficients of 0.37, 0.33 and 0.17 for OPER, COUP and AMIP respectively. In contrast to the monthly mean distributions, the agreement of the daily averages of the assimilated model versions with observations is significantly better than that of the non-assimilated model.

The agreement between SO_2 and SO_4^{2-} surface concentrations with observations over Europe and Canada was worse than that for precipitation. Predicted SO_2 concentrations at most sites were more than a factor of two higher than the observations. SO_4^{2-} is calculated to within a factor of two of the observed values at 76% (OPER) and 38% (COUP) of the EMEP stations and at all CAPMON stations. Due to a lack of an actual emission database we used emission data valid for autumn 1985. Emissions are known to have declined between 1985 and 1993, particularly in Germany where 22 of 34 sites used reported a factor-of-two reduction in sulphur emissions. We clearly need a better emission database, which is not only up to date but also reflects the spatial and temporal variability of the sulphur emissions, at least over the region where the evaluation with observations is performed. In contrast to the absolute concentrations, the day-to-day variability is captured much better by the model. The observed and calculated time series consisting of 30 daily mean mixing ratios of SO_2 are significantly correlated at the 95% confidence level at 49% and 46% of the EMEP sites and at 40% and 60% at the CAPMON sites for OPER and COUP, respectively. SO_4^{2-} mixing ratios correlate at the 95% confidence level at 40% and 79% at the EMEP sites and at 50% and 60% at the CAPMON sites for OPER and COUP, respectively. To summarize: the assimilated model version is able to reproduce daily mean precipitation fluxes quite well; additionally, it is able to predict the temporal variability of transport and wet removal processes connected to the cyclonic activity in midlatitudes. The correlation for SO_4^{2-} is significantly improved if interactions between cloud physics and aerosol physics, as in COUP, are considered.

Finally, we have compared model predictions of various quantities with aircraft measurements taken off the coast of Nova Scotia. We find a satisfying agreement of
variables like wind, temperature and relative humidity which have been nudged to ECMWF data with aircraft observations. Variables, which show a high temporal and spatial variability, like cloud liquid water and sulphur concentrations are in poorer agreement with observations. A feature nicely captured by the model is that $\text{SO}_4^{2-}$ maxima are often found above maxima of in-cloud occurrence, indicating that most of the $\text{SO}_4^{2-}$ is formed in clouds. However, the prediction of point values which are not averaged either spatially or temporally is beyond the scope of such a model. Due to an insufficiently accurate emission database and the large uncertainty in observed cloud parameters and sulphur species, it is hard to judge which experimental design performs best in terms of clouds and sulphur species. OPER predicts the precipitation more realistically; COUP, which allows a feedback between the calculated sulphate aerosols and the cloud microphysics, performs better in terms of $\text{SO}_4^{2-}$ distribution. We can only conclude that the first results of this assimilated model version are encouraging, and the technique will be applied further to evaluate different model aspects in more detail. In addition, such model results provide useful information for off-line chemical-transport models, which are not included in other databases, like liquid water content or precipitation formation rates.

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