SPIDER model process studies of aircraft plume dilution using simplified chemistry

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ABSTRACT: The box model SPIDER was developed to test and compare various approaches to include the effect of aircraft plume processes (effective emissions indices) in large scale chemistry transport models and climate-chemistry models. Its simplified NO_x-O_3 chemistry parameterises only the most relevant non-linear processes. SPIDER reproduces the main features of more sophisticated plume models. Multi-plume interactions illustrate the capability of the SPIDER model.

1 MOTIVATION

Emissions from aircraft impact on global climate (cf. Brasseur et al., 1998; IPCC, 1999; Sausen et al., 2005). They are usually implemented in General Circulation Models (CGM) or Chemistry Transport Models (CTM) by an instantaneous dispersion of the emitted matter over the large-scale grid boxes. Following Petry et al. (1998), this is called the instantaneous dispersion (ID) approach. The ID approach neglects non-linear chemical conversion processes in the evolving single plume. To resolve these by a plume model is called the single plume, or SP approach. However, detailed SP chemical modelling is computationally too demanding, both for more complex principle studies of plume-plume interaction in a grid box, and for operational implementation in large-scale models.

For improvement of the ID approach in GCMs, Effective Emission Indices (EEIs) can be used (e.g., Möllhoff, 1996; Petry et al., 1998). These, and several other approaches to the problem, e.g., by Meijer et al. (1997), Karol et al. (1997, 2000), Kraabøl et al. (2000) and Kraabøl and Stordal (2000) all applied detailed chemistry schemes, while a simplified model to test and compare the various EEI concepts, and to perform studies of multi-plume interactions remains desirable.

The present paper reports on the development of such a box model with simplified chemistry, the SPIDER (SP-ID Emission Relations) model. The following sections focus on setup of the model, validation and first application to a plume-plume interaction. Further model development and applications will be described in a forthcoming paper.

Motivated by the work by Petry et al. (1998) who applied a detailed chemistry scheme, we aim at computing plume dilution, and comparing of ID and SP results using a computationally efficient box model with greatly simplified chemistry. The resulting SPIDER model avoids explicit solution of the chemical rate equations. Chemistry enters the equations only in parameterized form by "dynamic forcing" terms, and the only species considered are NO_x and O_3.

The objectives are to apply the validated SPIDER model to more complex cases, e.g. multiple plumes or the interaction between neighbouring GCM grid cell NO_x or O_3 fields, and to eventually evaluate different EEI approaches.

1.1 *SPIDER model setup*

The main process to be covered by the model is the non-linear production of ozone by aircraft NO_x emissions at cruise altitude. Hence, the first simplification in the SPIDER system of equations is to include only these two species: NO_x and O_3.

The physical processes which are to be explicitly included in and resolved by the model within a typical GCM grid box volume are a) the emission of NO_x inside the GCM box, S_{NOx}, b) non-linear production of ozone, P_{O3}, and c) the decay of the NO_x and O_3 fields by conversion to reservoir spe-

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cies. For treatment of the SP approach, additionally the background (outer domain, superscript o) and plume fields (inner domain, superscript i) have to be integrated separately, and the entrainment of background matter by turbulent mixing at the growing-plume boundary enters as another individual term in the budget equations.

1.1.1 *ID budget equations*

In Eqs. (1-4), the following units hold $[NO_x]$ = mol, $[O_3]$ = mol, $[no_x]$ = nmol mol^{-1}, following the well-known convention to denote extensive quantities by upper-case, and intensive quantities by lower-case letters:

$$d_t\, NO_x = S_{NOx}\; \delta(t-t') - \frac{1}{\tau_{NOx}} NO_x \quad , \tag{1}$$

$$d_t\, O_3 = P_{O3}(no_x) \qquad - \frac{1}{\tau_{O3}} O_3 \quad . \tag{2}$$

The reference background state without aircraft emissions follows for $S \equiv 0$, and d_t denotes the temporal derivative *d/dt*.

The decay, or conversion of NO_x and O_3 to reservoir species, is modeled as an exponential decay with fixed half-time periods τ (τ_{NOx} = 10 days, τ_{O3} = 30 days, cf. Köhler and Sausen, 1994). Future versions of SPIDER will include a typical diurnal variation of these time scales, but this is a second-order effect, and neglecting the diurnal cycle here has little consequence on the results.

1.1.2 *SP budget equations*

In Eqs. (3) and (4), each species must be treated with one budget equation for the plume (superscript i) and the background (superscript o). As the box model reference volume is one GCM grid box, the computation of entrainment, with a linear plume growth rate, in Eqs. (3-4) is terminated as soon as the plume volume V^i is equal to the reference volume V_{GCM}.

$$d_t\, NO_x{}^i = S_{NOx}\; \delta(t-t') + NO_x{}^o / V^o\; d_t V^i - \frac{1}{\tau_{NOx}} NO_x{}^i \quad , \tag{3a}$$

$$d_t\, NO_x{}^o = \qquad - NO_x{}^o / V^o\; d_t V^i - \frac{1}{\tau_{NOx}} NO_x{}^o \quad , \tag{3b}$$

$$d_t\, O_3{}^i = P_{O3}(no_x{}^i) \qquad + \; O_3{}^o / V^o\; d_t V^i - \frac{1}{\tau_{O3}} O_3{}^i \quad , \tag{4a}$$

$$d_t\, O_3{}^o = P_{O3}(no_x{}^o) \qquad - \; O_3{}^o / V^o\; d_t V^i - \frac{1}{\tau_{O3}} O_3{}^o \quad . \tag{4b}$$

Eq. (3a) allows including the case in which a fresh aircraft plume is emitted along the axis of an aged plume emitted by another aircraft earlier on. This case was already investigated by Kraabøl and Stordal (2000), and will also be treated here in Sec. 3.2

1.2 *Parameterisation of $P_{O3}(no_x)$ terms*

The non-linear production of ozone as a function of the ambient NO_x concentrations remains to be specified for the SPIDER model equations (2) and (4). As treated in detail by, e.g., Johnson and Rohrer (1995), Brasseur et al. (1996), Grooß et al. (1998), and Meilinger et al. (2001), the production of ozone does not only depend on NO_x concentrations, but is a highly variable function of other species like O_3 itself, H_2O, CO, hydrocarbons, state variables p, and T, and the actinic flux J. A perfect parameterisation in this multidimensional phase space is impossible, and likely has prevented earlier studies using simplified chemistry studies of aircraft plume dilution.

However, in the present context, the objective is to develop a model which allows for principle studies of plume dilution, plume interaction, and methods to derive EEIs. Hence, a parameterisation of ozone production as a function of nitrogen oxides for some typical atmospheric conditions at cruise altitude following the data presented in the literature is possible. Aside from the NO_x concentration, also the solar elevation angle must be taken into account, in order to capture the diurnal cycle of photochemical ozone production.

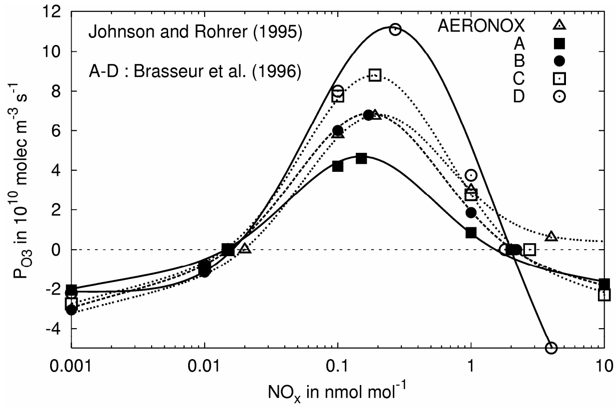

Figure 1. Net ozone production rate P_{O3} as a function of ambient NO_x concentration. The symbols are data from Johnson and Rohrer (1995) from the AERONOX project, and from Brasseur et al. (1996). The curves are fits to these data and form the selectable SPIDER P_{O3} parameterisations.

Figure 1 shows five different parameterisations of which D was selected in the SPIDER model. Curve D from the Brasseur et al. (1996) data includes effects of the diurnal cycle, the other curves are very similar in shape, and their variation comes mainly from different ambient chemical conditions.

Note the non-linearity, or rather non-monotonicity, of all P_{O3} curves. Low and very high NO_x concentrations are characterized by ozone depletion, while the peak ozone production is found in the range of 0.15 to 0.27 nmol mol^{-1}. The fact that the shape of the curves is quite uniform in the upper troposphere gives us some confidence that the SPIDER parameterisation of P_{O3} holds in a general sense and is adequate for principle process studies.

2 RESULTS

Here, we present the SPIDER model validation and its first application of multiple plume effects.

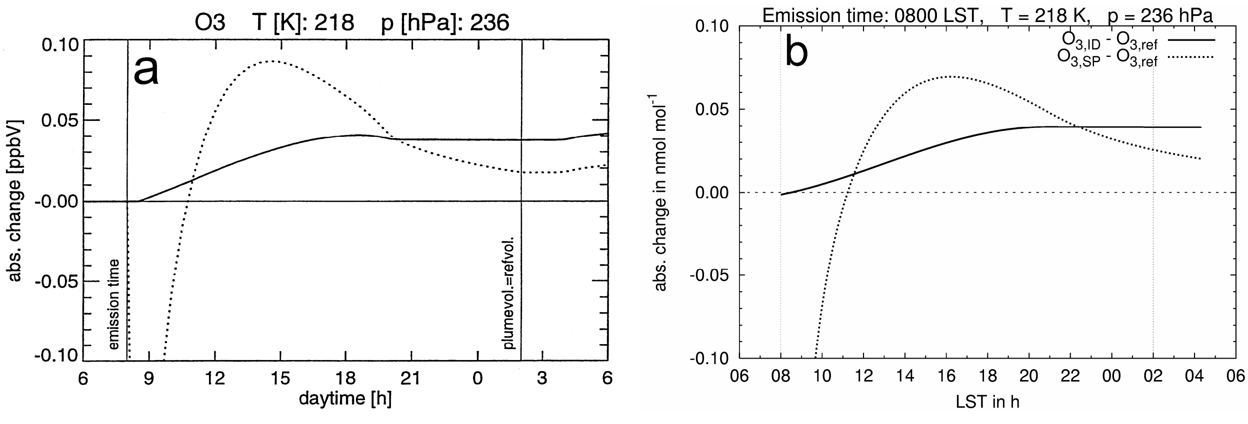

Figure 2. Absolute aircraft-induced change of O_3 concentration compared to the background state for ID (solid) and SP simulations (dotted). (a) is from Möllhoff (1996), (b) shows the corresponding SPIDER run.

2.1 *SPIDER model validation and sensitivity*

We used the original model cases from the work by Möllhoff (1996) to validate the SPIDER model. There, without wind shear or cross-plume wind components, the exhaust of a typical B747 airplane was emitted as a line-source at 0800 LST (local solar time) in a V_{GCM} = 50 x 50 x 1 km^3 reference volume. Ambient conditions were mid-latitude summer, T = 218 K and p = 236 hPa (about 10 km above see level, ASL). The initial values of NO_x and O_3 in the plume were chosen to be representative of the early dispersion regime (about 100 s after emission). Linear plume growth was specified such that after 18 h of plume dilution, the plume volume was equal to the reference volume V_{GCM}.

Figure 2b shows that the qualitative behaviour of the Möllhoff (1996) simulation in Figure 2a is captured well by the SPIDER model. The quantitative agreement is adequate; the main difference is that in the SP simulation, the peak change in O_3 is at a lower level and slightly later for the SPIDER run. For the ID run, the small peak before converging to the night time stable state is not resolved by SPIDER; instead, it merely converges towards the night time conditions.

The first few minutes after plume emission are characterised by ozone titration within the plume due to the very high NO_x concentrations (cf. Fig. 1, curve D). As the SPIDER model equations are formulated for the plume dispersion regime (the far-field solution), they cannot resolve the initial titration, which is a near-field plume process. Following Veenstra and Beck (1994), the initial ozone level in the plume must be lowered slightly compared to the background state to provide the proper initialisation values for the early dispersion regime[1].

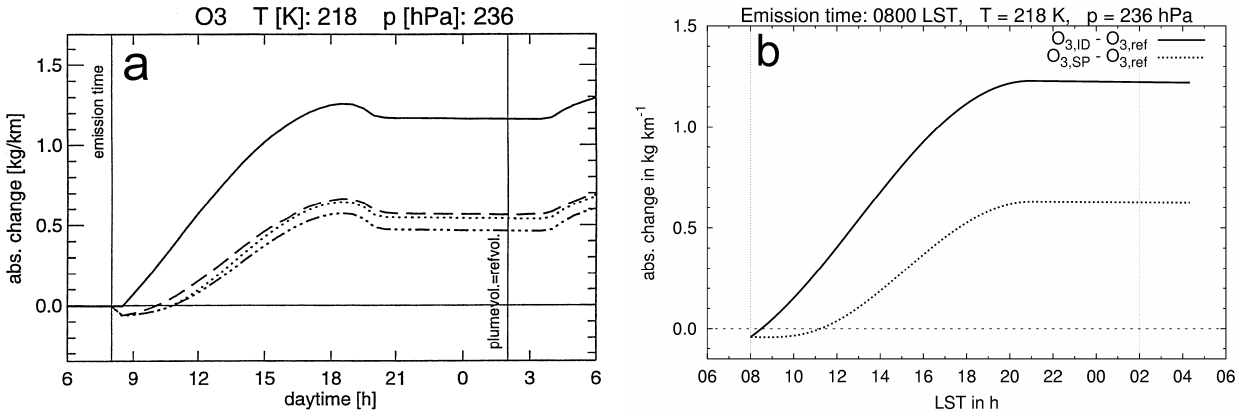

Figure 3. As Figure 2, but for the absolute change in O_3 mass per kilometre plume along the flight path. The dashed and dash-dotted lines in (a) from Möllhoff (1996) are not interpreted here.

Figure 3 shows the comparison between Möllhoff (1996) and the SPIDER results in terms of ozone mass difference per kilometre plume. Again, the small peak just before reaching the night time levels with zero photochemistry is not reproduced by SPIDER. Otherwise, the qualitative and quantitative agreement is good. Note that SPIDER correctly shows the extended period of negative change in ozone between 0800 and 1100 LST, and that the night time levels are well-captured.

2.2 *Aircraft following on track of initial one*

The first SPIDER model application case is a second, identical, aircraft exactly following the track of the first one after 4.5 h, injecting (and instantaneously distributing) a fresh plume into the aged, diluted one. For regions like the North Atlantic flight corridor, we consider this scenario to be quite realistic. Then, ID and SP simulations are continued and compared to the reference run. A similar case was investigated by Kraabøl and Stordal (2000), but for emission of the young plume 1 h after the first one (and additional runs for 2 h and 3 h release time lag).

Figure 4 shows the effects for the absolute change in ozone concentration (a) and mass per kilometre plume (b). The results should be compared to the reference case in Figs. 2b and 3b. The immediate effect of the young plume is visible in both panels by the initial drop in ozone due to titration. After recovery, the rate of ozone production is significantly enhanced, and higher night time levels of ozone result. For the ozone changes in Figure 4, the enhancement is about 50% for both the ID and SP runs, and for both the concentration and mass changes.

The response to the increase in aircraft NO_x is not linear here (in contrast to GCM simulations, cf. Grewe et al., 1999), as SPIDER experiences the full non-linearity of the P_{O3} term due to the initially high NO_x concentrations. I.e., the ozone production is not doubled by injection of the second plume after 4.5 h. Yet what can be said is that the gap between the results of the ID and SP approaches widens by roughly 50% due to the interaction of the two plumes. Thus, conclusions derived from single plume comparisons between SP and ID results, like that of Petry et al. (1998) stat-

[1] For completeness, we note that SPIDER is capable of reproducing initial ozone titration when the ozone concentration in the plume is initialised to the reference value. However, this titration is much weaker than that of Fig. 2

ing that for diurnal or seasonal averages, the difference between SP and ID plume dilution is not significant, may not remain justified in regions with frequent interaction of plumes.

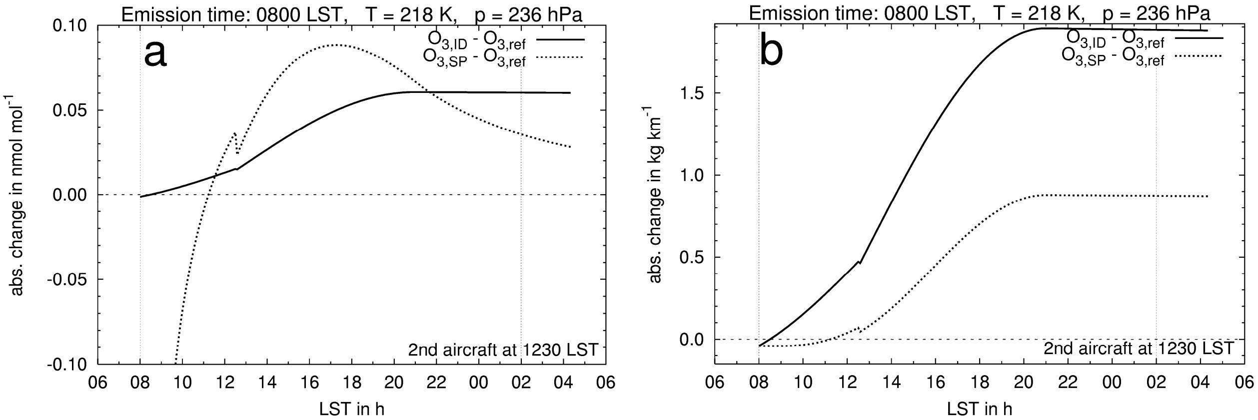

Figure 4. As Figs. 2b (a) and 3b (b), but with a second, identical aircraft plume emitted along the original flight path 4.5 h after emission of the first plume. Emission time 0800 LST is marked by the dotted line.

3 DISCUSSION

The SPIDER model could be validated in the framework of its simplifying assumptions. The basic plume dilution processes are well represented, in part even quantitatively. Some details are missing in the model which would require the complete set of chemical reactions – or an improved description of either the plume growth (being linear only on average, cf. Schumann et al., 1998) or the actinic flux in the P_{O3} term. These latter points are current work in SPIDER development.

In our simulation of interaction of two coaxial plumes, a net increase in produced ozone was found, and the gap between SP and ID approaches widened. Kraabøl and Stordal (2000) did a similar study, but for emission of the young plume 1 h after the old one. They employed a full chemistry model, slightly different release time (0700 LST), and other initial values of plume and background NO_x and O_3 concentrations. In contrast to our findings, they reported a net decrease in ozone compared to the single-plume run, and less conversion of the emitted NO_x. Qualitatively similar results were reported for secondary plume release after time lags of 2 h and 3 h, respectively.

There may be several reasons for these differences to our SPIDER results. Kraabøl and Stordal (2000) emitted their second model plume in ambient conditions characterised by very high NO_x concentrations (6.8 nmol mol^{-1}). Without further debate on the realism of these high NO_x values for 1 to 3 h old plumes, their young plume experienced conditions of strong ozone titration (cf. Fig. 1). In the SPIDER model run 4.5 h after emission of the first plume, the aged plume NO_x concentration is well below 2 nmol mol^{-1}, i.e., definitely in a region of the P_{O3} term of Figure 1 with at least weak ozone production. Besides, Kraabøl and Stordal (2000) compared their plume concentrations with background conditions which changed with time on injection of the second plume. In the present SPIDER case, the background was not altered after introduction of the young plume, in order to be able to compare the results to the single-plume reference run.

4 CONCLUSIONS

Our study using the SPIDER box model showed:

- The model is well-suited for principle studies, and could be validated qualitatively and in part quantitatively using results by Möllhoff (1996) and Petry et al. (1998);
- The model reproduces the high sensitivity to plume and background NO_x and O_3 concentrations;
- Multi-plume interactions can alter the gap between of ID and SP plume dilution approaches.

Aside from more complex multi-plume and GCM grid box interactions, future work will encompass refinement of the computation of plume growth, actinic flux, and NO_x or O_3 decay times.

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