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SPIDER model process studies of aircraft plume dilution using simplified chemistry

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Abstract

To include the effect of aircraft plume processes in large-scale chemistry transport models and climate-chemistry models by effective emission indices, the instantaneous dispersion (ID) and single-plume (SD) approaches exist. The box model SPIDER was developed to evaluate these two concepts. 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 and the consideration of varying NO_x background concentrations illustrate the capability of the SPIDER model. Simulations for varying NO_x background reveal the largest difference between the ID and SP approaches in clean-air conditions. For a NO_x background of ~0.2 nmol mol^{-1}, the ID and SP approaches result in aviation-induced O_3 changes of opposite signs. Hence, this transition regime may require more attention in plume parameterisations applied in global atmospheric models.

Keywords: Aircraft emissions; Plume dilution; Box model; Simplified chemistry; Effective emission indices

SPIDER-Modell Prozess-Studien der Flugzeugabgasausbreitung mit vereinfachter Chemie

Zusammenfassung. Um Auswirkungen von Prozessen in Flugzeugabgas-fahnen in großskaligen Chemietransport- und Klima-Chemie-Modellen mittels effektiver Emissionsindizes zu beschreiben, existieren die Methoden der instantanen Verteilung (ID) und der „einzelnen Abgasfahne“ (SP). Das Box-Modell SPIDER wurde zur Evaluierung diese beiden Konzepte entwickelt. Seine vereinfachte NO_x-O_3 Chemie parametrisiert nur die wichtigsten nichtlinearen Prozesse. SPIDER reproduziert die Haupt-eigenschaften höher entwickelter Abgasfahnenmodelle. Die Wechsel-wirkung mehrerer Abgasfahnen und die Betrachtung variierender NO_x Hintergrundkonzentration veranschaulichen die Fähigkeiten des SPIDER

Modells. Simulationen mit variablem NO_x Hintergrund liefern die größten Unterschiede zwischen den ID und SP Herangehensweisen in Klarluft-Bedingungen. Für einen NO_x Hintergrund von ~0.2 nmol mol^{-1} führen die ID und SP Verfahren zu Luftfahrt-verursachten O_3 Änderungen von unterschiedlichem Vorzeichen. Daher könnte es nötig sein, diesen Übergangsbereich in Abgasfahnen-Parametrisierungen für globale Atmosphärenmodelle stärker zu beachten.

Schlagworte: Flugzeugemissionen; Abgasfahnenausbreitung; Box-Modell; vereinfachte Chemie; Effektive Emissionsindizes

1 Introduction

Emissions from aircraft have an 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, or ID approach. The ID approach neglects non-linear chemical conversion processes in the evolving aircraft exhaust plume. To resolve these by a plume model is called the single plume, or SP approach. However, detailed SP chemical modelling is computationally demanding, both for more complex principle studies of plume-plume interaction in a grid box, and for operational implementation in large-scale models.

To improve the ID approach in GCMs, Effective Emission Indices (EEI) can be used (e.g., MÖLLHOFF, 1996; PETRY et al., 1998). These, and several other approaches to the problem, for example, by MEIJER et al. (1997), MEIJER (2001), KAROL et al. (1997, 2000), KRAABØL et al. (2000), KRAABØL and STORDAL (2000), ESLER (2003), ESLER et al. (2004), and FRANKE et al. (2008) applied detailed chemistry schemes. Yet, a simplified scheme to test 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, DOTZEK and SAUSEN, 2007) model. The following sections first focus on setup of the model, validation and application to a plume-plume interaction. Then, this paper aims (1) to apply the SPIDER model to various NO_x backgrounds and (2) to identify those NO_x background concentrations where the application of a more sophisticated SP approach yields results significantly different from the ID approach. In Sec. 2, the model design is outlined. Sec. 3 presents the results, while Secs. 4 and 5 provide discussion and conclusions.

2 Model design

Motivated by the work by MÖLLHOFF (1996) and PETRY et al. (1998) who applied a detailed chemistry scheme, we aim at computing plume dilution, and comparing 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 parameterised 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, for instance, multiple plumes or varying background NO_x fields, and to eventually evaluate different EEI approaches.

2.1 SPIDER model setup

The main chemical process to be covered by the model is the non-linear production of O_3 by aircraft NO_x emissions at cruise altitude. Hence, the SPIDER system of equations includes only these two species.

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, E_{NOx}, b) the non-linear production of ozone, P_{O3}, and c) the decay of NO_x and O_3 fields by conversion to reservoir species. For treatment of the SP approach, 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 the budget equations as another individual term. The numerical treatment of the entrainment has been implemented with a very accurate scheme to ensure mass conservation of the species at all times.

As the SPIDER model equations are formulated for the plume dispersion regime (the far-field solution), they do not resolve initial titration, which is a near-field plume process. Following VEENSTRA and BECK (1994), the initial O_3 level in the plume must be lowered slightly compared to the background state to provide the proper initialisation values for the early dispersion regime. Eqs. (1-4) specify the budget equations for the ID and SP concepts. Following the convention, we denote extensive quantities by upper-case ($[NO_x]$ = mol, $[O_3]$ = mol) and intensive quantities by lower-case letters ($[no_x]$ = 10^{-9} = nmol mol^{-1}). The parameterisation of photochemical O_3 production P_{O3} applied to both approaches is presented in Sec. 2.2.

2.1.1 ID budget equations

In the ID budget equations,

$$\frac{d}{dt} NO_x = E_{NOx}\,\delta(t-t') - \frac{1}{\tau_{NOx}} NO_x \quad , \tag{1}$$

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

the reference background state without aircraft emissions follows for $E \equiv 0$, and emissions from more than one aircraft can be included by adding emission terms, each with individual emissions *E* and emission times *t'*.

The 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 our results.

2.1.2 SP budget equations

In the SP budget 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 (second term on the r.h.s) with a linear plume growth rate is terminated as soon as the plume volume $V^{(i)}$ is equal to the reference volume V_{GCM}.

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

$$\frac{d}{dt} NO_x^{(o)} = \qquad - NO_x^{(o)} / V^{(o)}\, \frac{d}{dt} V^{(i)} - \frac{1}{\tau_{NOx}} NO_x^{(o)} \quad , \tag{3b}$$

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

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

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 as well in Sec. 3.2.

2.2 Parameterisation of $P_{O3}(no_x)$ terms

The non-linear production of O_3 as a function of the ambient NO_x concentrations remains to be specified for the SPIDER model Eqs. (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 O_3 does not only depend on NO_x concentrations, but is a highly variable function of other species like O_3 itself, H_2O, OH, HNO_3, 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, the objective in developing the SPIDER model was to allow for basic studies of plume dilution, plume interaction, and methods to derive EEIs. Hence, a parameterisation of O_3 production as a function of nitrogen oxides for some typical atmospheric conditions at cruise altitude following published observational data 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 O_3 production.

Fig. 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, as well as very high, NO_x concentrations are characterized by O_3 depletion, while the peak O_3 production is found for NO_x mixing ratios 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 confidence that the SPIDER parameterisation of P_{O3} holds in a general sense and is adequate for fundamental process studies.

2.3 Experimental model setup

We perform case studies of O_3 formation by an aircraft plume at cruising altitude. This setting is similar to the original model cases of MÖLLHOFF (1996) and was also used by DOTZEK and SAUSEN (2007) to validate the SPIDER model. Without wind shear or cross-plume wind components, the exhaust of a typical B747 airplane is emitted as a line-source at 0800 LST (local solar time) in a V_{GCM} = 50 km x 50 km x 1 km reference volume. Ambient conditions are mid-latitude summer, T = 218 K and p = 236 hPa (about 10 km above sea level, ASL) in the North Atlantic flight corridor. The initial average values of NO_x and O_3 in the plume are chosen to be representative of the early dispersion regime (about 100 s after emission): for NO_x 2.97 nmol mol^{-1} and for O_3 196.5 nmol mol^{-1} (PETRY et al., 1998). Linear Gaussian plume growth is specified, so after t_{ref} = 18 h of dilution, the plume volume $V^{(i)}(t)$ becomes equal to the reference volume V_{GCM}.

3 Results

To compare the ID and SP approaches for the modelled cases, the temporal evolution of aircraft-induced O_3 change is both presented as change in concentration and change in mass per kilometre plume length along the flight path.

3.1 SPIDER model validation and sensitivity

DOTZEK and SAUSEN (2007) used the original model cases from MÖLLHOFF (1996) to validate the SPIDER model. Fig. 2b shows that the qualitative behaviour of the MÖLLHOFF (1996) simulation in Fig. 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 lower and occurs 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 O_3 titration within the plume due to the very high NO_x (mainly NO) concentrations (cf. Fig. 1, curve D). As stated in Sec. 2.1, the initial O_3 level in the plume had to be lowered slightly compared to the background state to provide the proper initialisation values for the early dispersion regime[1].

Fig. 3 shows the comparison between MÖLLHOFF (1996) and the SPIDER results in terms of O_3 mass difference per kilometre plume. Again, the small peak just before reaching the night time levels without 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 O_3 between 0800 and 1100 LST, and that the night time levels are well-captured.

3.2 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), yet for emission of the young plume 1 h after the first one (and additional runs for 2 h and 3 h release time lag).

Fig. 4 shows the resulting effects for the change in O_3 concentration (a) and O_3 mass per kilometre plume (b). The results can 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 O_3 due to titration. After recovery, the rate of O_3 production is significantly enhanced and higher night time levels of O_3 result. For the O_3 changes in Fig. 4, the enhancement compared to Figs. 2b and 3b 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. That is, the O_3 production is not doubled by injection of the second, identical 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 comparisons between SP and ID results, like that of PETRY et al. (1998) stating 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.

3.3 Quantitative SP-IP comparison for various background NO_x concentrations

SPIDER model runs were performed for NO_x background concentrations of 0.05, 0.075, 0.1, 0.2, 0.5, 1, 2 and 3 nmol mol^{-1}, respectively. These cover the range from clean-air to strongly polluted environments. Fig. 5 shows exemplary results for 0.05, 0.1, 0.2 and 1 nmol mol^{-1} NO_x backgrounds. During the first few minutes after plume emission, aircraft-induced O_3 change is again characterised by O_3 titration within the plume due to very high NO_x concentration under all NO_x background conditions.

Both the change in O_3 concentration and the change in O_3 mass per kilometre flight path reveal that the largest differences between the ID and SP approaches materialise for clean-air ambient conditions, that is, for NO_x background concentrations of less than about 0.1 nmol mol^{-1}. After an initial O_3 titration in SP simulations, O_3 production during the early phase (up to several hours) is higher in SP simulations, compared to ID calculations. More than 12 hours after emissions, this changes, and finally O_3 production in SP simulations is lower than in ID calculations.

[1] Note that SPIDER is capable of reproducing initial O_3 titration even if the O_3 concentration in the plume is initialised to the background value. However, this titration is much weaker than that of Fig. 2.

For strongly polluted environments (1, 2, 3 nmol mol^{-1}, latter two not shown), the ID and SP simulations yield essentially identical results at the time when the plume attains the same volume as the GCM grid box. Interestingly, a transition regime can be identified for NO_x backgrounds of about 0.2 nmol mol^{-1}, in which the O_3 productions of the ID and SP approaches at $t = t_{ref}$ are small, but have opposite signs. In this regime, during 24 hours after emission, the SP approach leads to a small net destruction of O_3, while the ID approach leads to O_3 production with a magnitude considerably larger than the destruction evaluated from the SP approach. This opposite sign of O_3 change between the SP and ID approaches prevails from 4 hours after emission onward.

4 Discussion

The SPIDER model was successfully validated and applied in the framework of its simplifying assumptions. The basic plume dilution processes are well represented, in part even quantitatively. In our simulation of interaction of two coaxial plumes, a net increase in produced O_3 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 O_3 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 O_3 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}, that is, definitely in a region of the P_{O3} term of Fig. 1 with at least weak O_3 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.

Depending on NO_x background concentrations, substantial differences between the ID and SP approaches can occur. Differences in O_3 change observed in our results indicate that in the clean-air regime (below 0.1 nmol mol^{-1}) both ID and SP ozone productions are positive and show their largest absolute spreads. In the transition regime (~0.2 nmol mol^{-1}), an opposite sign can be observed between ID and SP approaches from several hours after emission onward. This pattern prevails even after 18 hours of plume expansion to occupy the GCM box volume. For more polluted regions, however, with NO_x backgrounds well above 0.2 nmol mol^{-1}, the ID and SP approaches yield increasingly similar results. Hence, for such conditions which can be found in the North Atlantic flight corridor, an ID approach may still be adequate and least time-consuming for application in GCMs or CTMs. Under clean-air conditions and in the transition regime, use of an ID approach would yield substantial differences from a more detailed SP approach, overestimating aviation-induced O_3 changes.

The simplifications made in the SPIDER model equations require more discussion. The basic plume dilution processes were shown to be well-represented by DOTZEK and SAUSEN (2007), 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. Nonlinear plume growth already has been implemented as an option in the model, but to facilitate comparison to the DOTZEK and SAUSEN (2007) results, it was not considered here. Our model set-up does not explicitly include a typical diurnal cycle. For the parametric functions of P_{O3}, a curve was selected from BRASSEUR et al. (1996) including a diurnal cycle. Future SPIDER versions will also include a typical diurnal variation of the conversion time scales to reservoir species, but this is a second-order effect with little consequence on the present results.

For multi-plume interactions, the net effect on the difference between the ID and SP approaches critically depends on the age of the primary plume and hence its NO_x and O_3 concentrations. However, our additional study with NO_x background variations across the whole GCM grid box showed a consistent trend. The need for a more sophisticated description of plume processes in GCMs sets in at NO_x backgrounds of about 0.2 nmol mol^{-1}, first with a disparity of the signs of the (small) O_3 productions, and then with increasing O_3 overestimates by the ID approach for less polluted regions.

The inclusion of plume effects in the dispersion modelling of pollutants is not only relevant in aviation at cruise altitude, but also near the ground (e.g., UPHOFF, 2008; GALMARINI et al., 2008), for land transport (e.g., GANEV et al., 2008) and shipping (e.g.,

CHOSSON et al., 2008; FRANKE et al., 2008; CHARLTON-PEREZ et al., 2009; KIM et al., 2009). Several parameterisations to include these effects in mesoscale or general circulation models have been proposed recently. CARIOLLE et al. (2009) specifically addressed aircraft NO_x emissions in a similar setting as in out present paper. They tracked the plume air with NO_x concentrations above 1 nmol mol^{-1} by introducing a "fuel tracer" and a characteristic lifetime in their budget equations. Their detailed parameterisation confirms our results: Taking into account the plume processes consistently lowers the estimates of aircraft-induced O_3 production at cruise altitude in parts of the North Atlantic flight corridor.

5 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 known from observations and detailed chemistry simulations;
- Multi-plume interactions can alter the gap between the ID and SP plume dilution approaches in a non-linear fashion;
- The largest differences between the ID and SP approaches occur for clean-air ambient conditions, that is, for NO_x background concentrations of less than ~0.1 nmol mol^{-1};
- For strongly polluted environments, the ID and SP simulations yield essentially identical results at the time when the plume attains the same volume as the GCM grid box;
- A transition regime can be identified for NO_x backgrounds of ~0.2 nmol mol^{-1} in which the O_3 productions of the ID and SP approaches after 18 h, albeit small, have opposite signs.
- It appears necessary to also consider this transition regime in parameterisations of the O_3 production by aircraft NO_x emissions at cruise altitude, in addition to the clean-air regime.

Aside from more complex multi-plume and GCM grid box interactions, future work could encompass refinement of the computation of plume growth, actinic flux, and NO_x or O_3 decay times, as well as simulations for a wider range of likely environmental conditions at cruise altitude to assess the robustness of our findings.

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Figure captions

Fig. 1: Net O_3 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.

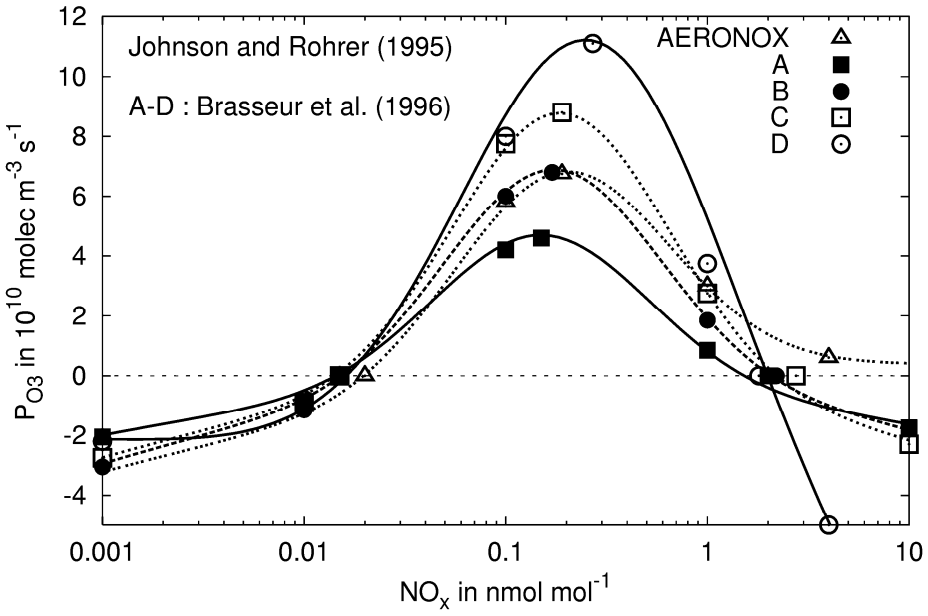
Fig. 2: Aircraft-induced change of O_3 concentration compared to the background state for ID (solid) and SP simulations (dotted): (a) is from MÖLLHOFF (1996), (b) shows the corresponding SPIDER run.

Fig. 3: As Fig. 2, but for the 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.

Fig. 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.

Fig. 5: Aircraft-induced O_3 concentration change (left panels) and change of O_3 mass per kilometre plume length along the flight path (right panels) compared to the background state for ID (solid) and SP simulations (dotted). Background NO_x concentrations increase from top to bottom: (a, b) 0.05 nmol mol^{-1}, (c, d) 0.1 nmol mol^{-1}, (e, f) 0.2 nmol mol^{-1}, and (g, h) 1 nmol mol^{-1}, Emission time was 0800 LST and after 18 h, the plume volume equals the GCM box volume (dashed line).

Figures



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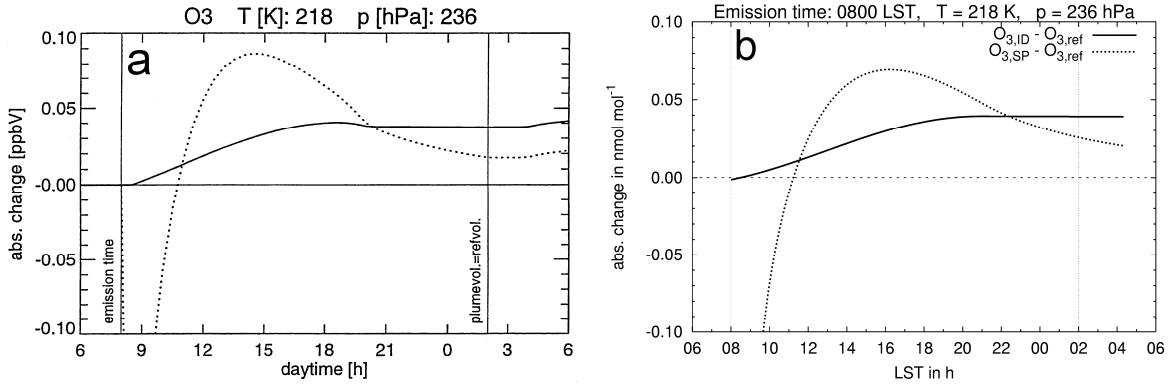

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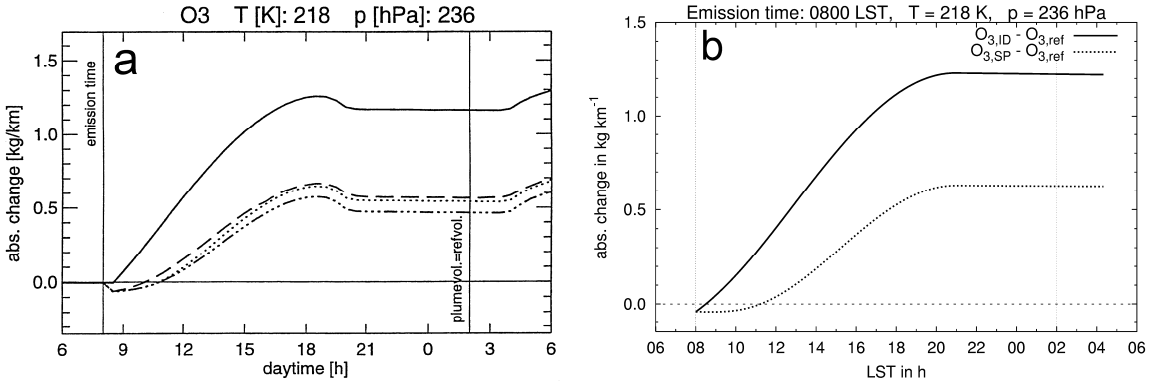

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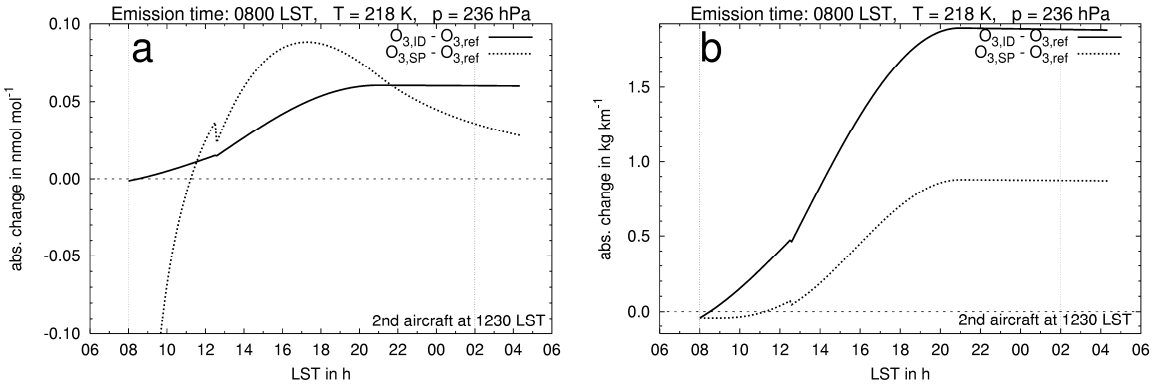

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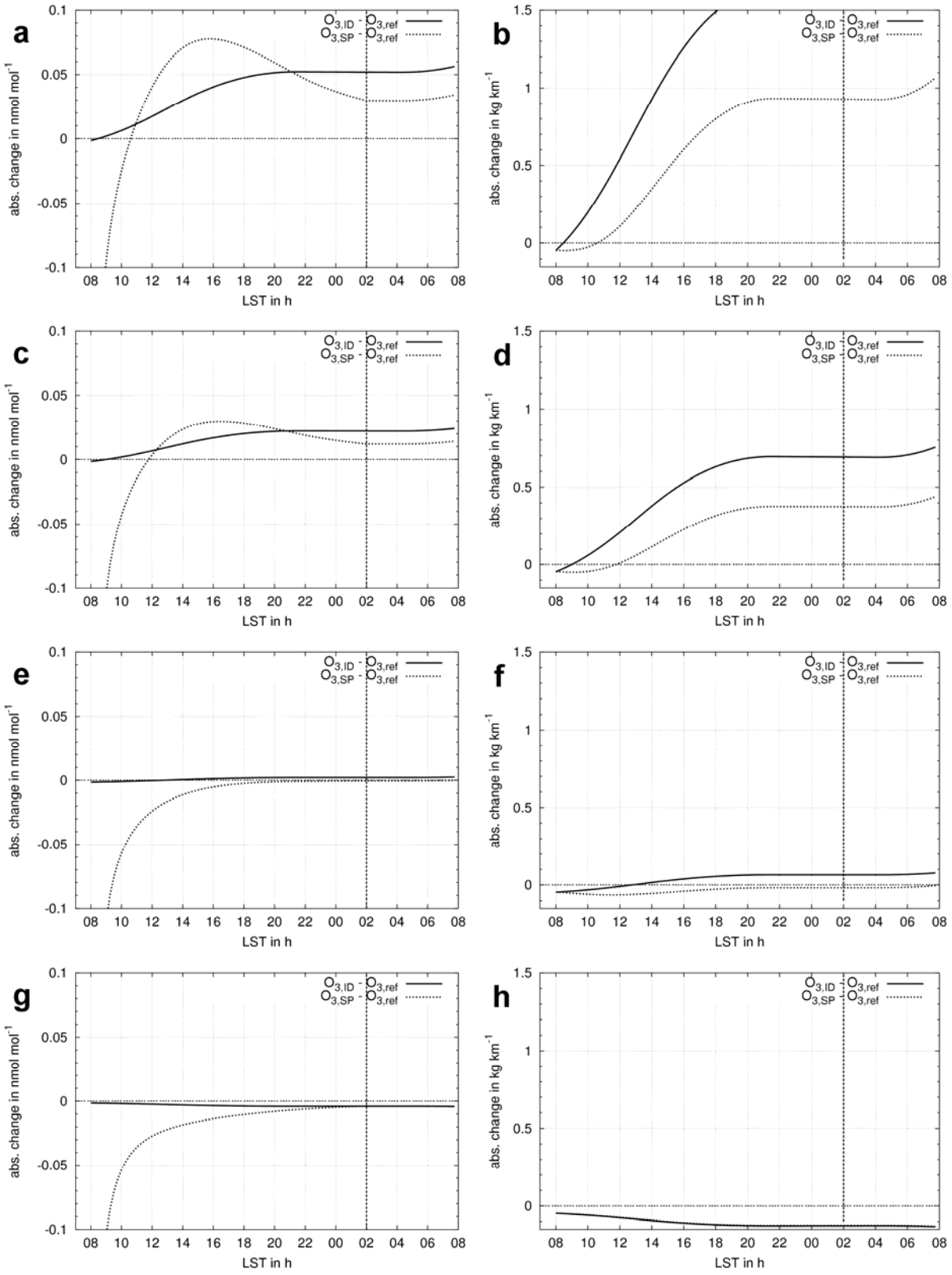

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