1 Comment 1 by Dennis Baldocchi

1.1 Comment from Referee

The field of dry deposition has had periods of ups and downs in activity and research. Unfortunately algorithms in important models have been fossilized to consider the Wesely model of 1989. While that was a very good and appropriate algorithm 30 years ago, we know more about land surface fluxes, how to model stomatal conductance and have been datasets and parameterization information in 2020. So, I was excited to see this paper.
I see the main contributions are
The default dry deposition scheme has been extended with adjustment factors to predict stomatal responses to temperature and vapour pressure deficit. Furthermore, an explicit formulation of the non-stomatal deposition to the leaf surface (cuticle) dependent on humidity has been implemented based on established schemes. Finally, the soil moisture availability function for plants has been revised to be consistent with the simple hydrological model available in EMAC.
The authors make a good case for this work and its significance as 'the revision of the process parameterisation as documented here has the potential to significantly reduce the overestimation of tropospheric ozone in global models'.
This paper is a steps in the right direction, but revolves around the over parameterized Jarvis stomatal model that was used in the 80s with more adjustment factors. Many of us, including Piers Sellers, have abandoned the Jarvis model in land-surface modeling of water and carbon fluxes because it lead to stomatal suicide. Others have adopted the Ball-Berry approach, with better fidelity
Baldocchi, D. D., and T. Meyers (1998), On using eco-physiological, micrometeorological and biogeochemical theory to evaluate carbon dioxide, water vapor and trace gas fluxes over vegetation: a perspective, Agricultural and Forest Meteorology, 90(1-2), 1-25.
I don't view this 'new'model as an improvement by going back to the Jarvis model for stomatal conductance. There has been many advances in stomatal modeling worth considering in 2020.
Wang, Yujie, John S. Sperry, William RL Anderegg, Martin D. Venturas, and Anna T. Trugman. "A theoretical and empirical assessment of stomatal optimization modeling." New Phytologist (2020).

Medlyn, B. E., Duursma, R. A., Eamus, D., Ellsworth, D. S., Prentice, I. C., Barton, C. V., ... & Wingate, L. (2011). Reconciling the optimal and empirical approaches to modelling stomatal conductance. Global Change Biology, 17(6), 2134-2144.

Personally, I'd like to see some connection with ecosystem photosynthesis scaling with stomatal conductance. There has been excellent advances modeling both that could be coupled with a stomatal and dry deposition model, for instance.

Jiang, C., and Y. Ryu (2016), Multi-scale evaluation of global gross primary productivity and evapotranspiration products derived from Breathing Earth System Simulator (BESS), Remote Sensing of Environment, 186, 528-547, doi:http://dx.doi.org/10.1016/j.rse.2016.08.030.

De Kauwe, Martin G., et al. "A test of an optimal stomatal conductance scheme within the CABLE land surface model." Geoscientific Model Development (2015): 431-452.

In writing the introduction, there has been some recent workshops on dry deposition, newer long term studies and a very good review that should be cited and considered

Clifton, O. E., Fiore, A. M., Massman, W. J., Baublitz, C. B., Coyle, M., Emberson, L., ... (2020). Dry deposition of ozone over land: processes, measurement, and modeling. Reviews of Geophysics, 58(1), e2019RG000670.

Clifton, O. E., A. M. Fiore, J. W. Munger, S. Malyshev, L. W. Horowitz, E. Shevliakova, F. Paulot, L. T. Murray, and K. L. Griffin (2017), Interannual variability in ozone removal by a temperate deciduous forest, Geophysical Research Letters, 44(1), 542-552, doi:10.1002/2016gl070923.

Clifton, O. E., Paulot, F., Fiore, A. M., Horowitz, L. W., Correa, G., Baublitz, C. B., ... (2020). Influence of dynamic ozone dry deposition on ozone pollution. Journal of Geophysical Research: Atmospheres, 125(8), e2020JD032398.

I am of mixed feelings of this work. I find the model algorithm dated and not an improvement. On the other hand there has been a dearth of long term flux measurements and use of those data to test the performance of a model, as it done here.
To my opinion this would be much better paper by using modern, better state of art stomatal models that couple carbon and water fluxes and test the performance against a year of flux measurements. Then I would feel the work is new, novel and a significant improvement over the past work.
I also like the use of 4 contrasting flux datasets. This too is an advance in model testing. For example regarding performance, we learn 'As seen from the comparison of stomatal resistance values (Fig. 4d) the model underestimates the stomatal uptake. This is because the irrigation of the Orchard leads to cooling sustained evapotranspiration and keeps f(T) low.Thus in the model, a too high temperature stress act on the stomata'.
My alternative hypothesis is that this bias may disappear with a coupled carbon-water stomatal conductance model.
If I have learned anything over my career it is the power and importance of multiple constraints. Sadly, the Jarvis model does not deliver. It was great circa 1976 and helped us think about the role of stomata on dry deposition in the 1980s, but that is its extent of being good enough.

Fig 3 would be better if error bars were added, given these are monthly means. I do like the global upscaling. It helps address the 'so what?' question and does produce some multiple constraint with regards to getting pollution right, as we see in Fig. 6.
My bottom line is that this paper can be remedied. It has lots of strengths worth keeping. And the spirit of the work is good.
Regarding conclusion
The seasonal variability of the simulated dry deposition velocity could be further improved by using as model input the time-series of vegetation cover from an imaging products which also capture land use changes and vegetation trend that are known to impact dry deposition significantly.
Connection to phenology modeling or observation is key to getting the seasonality in LAI correct and the fluxes right. So Yes this is an important aspect of the model. I'd like to see it in the 'new model'.
If the model had already coupled water and carbon phenology should be part of it.

1.2 Author's response

Dear referee, many thanks for your review. Here are our replies: The article documents a revision of the existing dry deposition scheme in EMAC not a complete new implementation. The idea is to improve the existing scheme based on the already available information in the model (i.e. without detailed phenology information etc.) because model results show that a more precise representation could lower the overestimation of ozone by models. With the current model version, these developments can only draw on limited vegetation information without details on cover and phenology. Dry deposition of trace gases is represented by the "resistance-in-series" scheme of Wesely (1989). The stomatal uptake was firstly only based on the response to incoming solar radiation developed by Sellers (1985) which is known to be an important fluctuation factor (Dawson et al. 2010), and a soil moisture stress factor. The further developments were build on this common dry deposition scheme. For the extension with additional stress factors, we adopt the multiplicative principle and the temperature stress factor by Jarvis (1976). This principle is commonly used in second-generation LSM schemes due to its computational efficiency, adaptability and simplicity (Pitman et al. 2003, Clifton et al. 2020) and has been shown to capture 95 % of the observed variability of stomatal conductance (Dawson et al 2010). The stomatal sensitivity to vapour pressure deficit is calculated according to the optimisation framework by Katul et al. (2009) which maximises the use of carbon under a minimal cost of water inside the plant. This concept accounts for the water cost of carbon without specifying the stomatal response to VPD and CO_2 in advance and agrees well with experimental data (Katul et al. 2009). Hence, by adding also the stomatal response to temperature and vapour pressure deficit within this study, the key responses of stomates are represented (Pitman et al. 2003). Comparing to measurement data, several studies found that Jarvis-type models can compete with Ball-Berry models in explaining observed stomatal conductance and stomatal ozone flux to vegetation (Hoshika et al. 2017, Ran et al. 2017) whereas both have different limitations and advantages (Lu 2018, Farquhar et al. 1980). The performance of both models depend certainly on the choice of parameters (Sulis et al. 2015, Lu 2018). The mentioned "stomatal suicide" as major critique to the Jarvis model has been experienced in EMAC and is attributed to the lack of soil moisture storage in some regions. It is solved currently by adapting the soil moisture stress factor to the used soil representation. Moreover, the stress factor dependent on VPD (Katul et al. 2009), that we use, exerts a stronger control on evapostranspiration that the original factor proposed by Jarvis. For comparison, at VPD = 5 kPa stomatal conductance is predicted to decrease by about 50% and < 10% according to Katul et al. (2009) and Jarvis (1976), respectvely. A further amelioration of the EMAC model dry bias in the Amazon is brought by the use of VPD factor by Katul et al. (2009) only in simulations without meteorological nudging (not shown in the manuscript). The usage of the Ball-Berry approach is constrained by the availability of detailed information on plant microphysics which determine the parameters. Due to the current limitations of EMAC in this regard, described above, an implementation would build on many assumptions concerning the representation at global scale.

With regard to the developments of stomatal conductance models in the last years the approach used here is dated but in EMAC this represents a significant improvement compared to the existing parametrization.The adaptability, simplicity and computer efficiency makes it attractive for the use at global scale and the usage of parametrizations for radiation response and VPD stress are different from the one used in Jarvis (1976).

We agree with the Referee but unfortunately these developments are limited by the minimal ecosystem representation in the EMAC model. Implementing a mechanistic approach which connect stomatal conductance to plant photosynthesis is definitely intended for EMAC once a vegetation model with the sufficient details and well-constrained parameters will be available.

We will add a paragraph on the current research status of dry deposition to the introduction considering this studies .

With regard to the mentioned limitations and the current status of the dry deposition parametrization in EMAC, our development can be seen as an intermediate stage on the way to a "state-of-the-art" dry deposition scheme. For the stomatal part, major dependencies to meteorology have been established whereas the implementation of the cuticular pathway contributes to a global enhancement of dry deposition especially of soluble organic species that are ozone precursors. Furthermore, the study has a significance for the MESSy community as first technical description and evaluation of the vertical exchange submodel VERTEX.

Concerning the model evaluation at Citrus Orchard, we cannot exclude that such a model might remove the bias. However, if it did, it would do it for the wrong reasons. The absence of soil water stress at Citrus Orchard (due to irrigation) is artificial and not represented in the global model. Thus, the site cannot be representative for the mostly non irrigated 1.1°x1.1° grid box including Citrus Orchard. In fact, removal of the water stress from the model greatly reduces the model bias at Citrus Orchard (see Fig. 4d).

We are aware of the limitations of the implemented model parametrization. But regarding that the developments for a global model which has only a minimal ecosystem representation available, we see the current implementation as the best achievable in EMAC without having to embark on the coupling with a dynamic vegetation model that would provide the desired constraints.

Error bars can be added for all sub figures.

Thank you for mentioning this aspect which addresses the actual motivation of this model study. EMAC is an Atmospheric Chemistry Model which explicit chemistry and misses on the other hand details for e.g. the vegetation representation. Regarding all the arguments mentioned above we can not be sure that implementing a simple 'Anet-g_s' stomatal approach relying on the scanty vegetation information available in the model could improve the representation of dry deposition in EMAC.

The usage of the time-series of vegetation cover from the Moderate Resolution Imaging Spectroradiometer (MODIS) is in preparation as one of the few available means to represent ecosystem phenology in the current model. However, so far only LAI data from MODIS is available in the model and remaining data like canopy height still have to be acquired. Water and carbon phenology is unfortunately not yet part of the model and will be added as part of a future planned vegetation model for EMAC.

References

Yen-Sen Lu, Propagation of land surface model uncertainties in simulated terrestrial system states. 2018. PhD Thesis. Universitäts-und Landesbibliothek Bonn. http://hss.ulb.uni-bonn.de/2018/5071/5071.htm

Hoshika, Y.; Fares, S.; Savi, F.; Gruening, C.; Goded, I.; De Marco, A.; Sicard, P. & Paoletti, E. Stomatal conductance models for ozone risk assessment at canopy level in two Mediterranean evergreen forests. Agricultural and Forest Meteorology (2017), 234, 212-221.

Damour, Gaëlle, et al. "An overview of models of stomatal conductance at the leaf level." Plant, Cell & Environment 33.9 (2010): 1419-1438.

Farquhar, Graham D., S. von von Caemmerer, and J. A. Berry. "A biochemical model of photosynthetic CO 2 assimilation in leaves of C 3 species." Planta 149.1 (1980): 78-90.

Katul, Gabriel G., Sari Palmroth, and R. A. M. Oren. "Leaf stomatal responses to vapour pressure deficit under current and CO2-enriched atmosphere explained by the economics of gas exchange." Plant, Cell & Environment 32.8 (2009): 968-979.

Pitman, A. J. The evolution of, and revolution in, land surface schemes designed for climate models Int. J. Climatol., John Wiley & Sons, Ltd., 2003, 23, 479-510

Sulis, M.; Langensiepen, M.; Shrestha, P.; Schickling, A.; Simmer, C. & Kollet, S. J. Evaluating the Influence of Plant-Specific Physiological Parameterizations on the Partitioning of Land Surface Energy Fluxes J. Hydrometeor, American Meteorological Society, 2015, 16, 517-533

Jarvis, P. G. "The interpretation of the variations in leaf water potential and stomatal conductance found in canopies in the field." Philosophical Transactions of the Royal Society of London. B, Biological Sciences 273.927 (1976): 593-610.

Wesely, Ml. "Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models." Atmospheric environment 23.6 (1989): 1293-1304.

Sellers, Piers J. "Canopy reflectance, photosynthesis and transpiration." International journal of remote sensing 6.8 (1985): 1335-1372.

1.3 Author's changes in manuscript

- In Section 1 (line 34-46), the following was added:

> *Thereby, stomatal uptake is commonly parameterised following the empirical multiplicative approach by Jarvis (1976) which uses a predefined minimum resistance and multiple environmental response factors like in Zhang et al. (2003), Simpson et al. (2012) and Emberson et al. (2000). More advanced formulations often used by land surface models (Ran et al., 2017; Val Martin et al., 2014) are based on the CO_2 assimilation by plants during photosynthesis (Ball et al., 1987, Collatz et al., 1997). Both approaches rely on the choice and constraints of ecosystem dependent parameters and have different advantages (Lu, 2018). A further role in coupling stomata to ecosystems play stomatal optimization models whereas optimal stomatal activity with a maximum amount of carbon gain and a minimum loss of water is calculated based on eco-physiological processes (e.g. Cowan and Farquar, 1977). Of particular interest are stomatal optimization models which, based on eco-physiological processes, maximize carbon gain while minimizing water loss. According to Wang et al. (2020) these models are promising in representing stomatal behaviour and improving carbon cycle modelling. Non-stomatal deposition has been less investigated by now therefore most models use predefined constant resistances or scale it with leaf area index (e.g. Val Martin et al., 2014; Simpson et al., 2012) while some apply an explicit parametrization based on the observational*

findings of enhance cuticular uptake under leaf surface wetness (Altimir et al., 2006).

- Section 6 'Uncertainties in modelling stomatal conductance' was added.
- Error bars were added to Figure 3c-f

2 Comment 2

2.1 Comment from Referee

General Comments
Even there is always a severe lack of direct flux measurement, the sporadic efforts over the past 20 years still reveals a lot of new and interesting environment dependence and inter-site variabilities of gaseous dry deposition after the proposal of the ever-popular (Hardacre et al., 2015) Wesely scheme (Wesely, 1989) and its slight variants (e.g. Wang et al., 1998). Meanwhile, enormous advance has been made over modelling carbon-water exchange, and therefore stomatal modelling. And given that dry deposition has been shown as one of the major uncertainty of modelling surface ozone (Wong et al., 2019), therefore, I largely agree with the position of the first reviewer, that the effort of updating gaseous dry deposition schemes shall be welcomed and encouraged.
Yet, I doubt whether this paper is doing a good enough job in "updating" the dry deposition scheme, particularly in terms of modelling canopy resistance. Given the functional diversity of plants on the Earth, I find one of the biggest weakness of the scheme presented in this paper is the lack of biome-dependence of both its stomatal and cuticular parameters, especially given that previous works have already addressed this issue (e.g. Emberson et al., 2013; Simpson et al., 2012; Zhang et al., 2003). There is also notable weakness in evaluation of the proposed scheme, but it is much easier to address.

Specific Comments
Starting from stomatal conductance. I agree with the authors, that the simplicity and effectiveness of Jarvis-type parameterizations have its place in atmospheric modelling. Yet this particular ecophysiological theory itself (Jarvis, 1976) only states that stomatal conductance has multiple simultaneous constraint (mathematically, $g_s = g_{max} \prod_{i}^{n_{constraints}} f_i(X_i), 0 \leq f_i \geq 1$al functional forms (i.e. the mathematical forms of fi) and parameters of all biomes over the world. It has been explicitly shown that improperly parameterized Jarvis-type model can lead to substantial bias (Fares et al., 2013).
Earlier works of updating dry deposition schemes with Jarvis-type stomatal sub-models (e.g. Simpson et al., 2012; Zhang et al., 2003) had already been assigning stomatal parameters to each individual biome. Though one may argue that they are neither backed empirically (improperly parameterized), they are probably still working better, especially for global modelling, than one single set

of stomatal parameters over all biomes. For example, Hoshika et al. (2018) empirically derive that gsmax (maximum stomatal conductance) can vary almost ten-folds across all biomes, and the optimal temperature of stomatal opening (Topt) generally increases as the mean annual air temperature. The Zhang and EMEP parameterizations stated above are able to qualitatively capture some features showing in Hoshika et al. (2018) (e.g. higher gsmax for broadleaf trees and crops than boreal forests, higher Topt for tropical than boreal biomes), giving them more creditability when applied regionally and globally, which cannot be achieved by one single set of stomatal parameters applied to all biomes over the world. In fact, the large model-observation mismatch over ATTO (fig. 5), which the authors attribute to underestimated stomatal uptake (line 327), may also be a product improper parameterization more than inaccurate meteorology. The same problem happens similarly, but to a lesser extent, for the cuticular parameterization, as Zhang et al. (2003) did assign different cuticular uptake parameters for different land types. But it is much more difficult to assess whether these parameters make sense than their stomatal counter parts. So this should be a minor issue. However, some discussions on the uncertainty and inter-biome variability of these parameters is important.

Another main issue is the model evaluation, which may also stem from the fact that the proposed scheme has no biome dependence. The model evaluation over the four sites is mostly specific and well-thought. However, in most recent work involves evaluating (Silva and Heald, 2018; Wong et al., 2019), developing (Clifton et al., 2020b; Lin et al., 2019) or reviewing (Clifton et al., 2020a) dry deposition schemes, extensive effort have been done to compile worldwide ozone dry deposition measurements to gauge the performance of ozone over different biomes. Most of the above works have publicized their compiled ozone deposition measurements. Adding another part of evaluation that focus on the performance over different land types is necessary in both establishing the credibility of the proposed scheme and identifying its potential weakness, especially given this is a global model.

As both the vertical transfer and canopy resistance schemes are modified, the update should affect not only O3, but all trace gases. It would be interesting to include a brief description on the changes in some other important trace gases (e.g. NO_2, SO_2, HNO_3).

Technical comments:

Line 106:

Let's refer to fig. 4 of Baldocchi et al. (1987). Linear scaling always produces lower resistance, and therefore higher uptake, than proper canopy scaling. Therefore linear scaling should overestimate uptake instead of underestimate.

Line 110:

More discussions and acknowledgements on proposed (e.g. Mészáros et al., 2009; Stella et al., 2019) and implemented (e.g. Clifton et al., 2020b) soil deposition schemes are need.

Line 192:

How is wetness and snow-covered fraction calculated? How is it related to LAI?

These should be clarified.
Line 235:
There are also other important long-term measurements (e.g. Blodgett Forest, Harvard Forest). Why do you choose these particular four data sets out of all available ozone flux measurements for detailed evaluation? Additional justification is needed.
Line 254:
Non-stomatal deposition does not only include cuticular, but also soil uptake. Other terminology (e.g. total cuticular conductance) shall be used in placed of non-stomatal conductance to avoid confusion and imprecision.

References

Baldocchi, D. D., Hicks, B. B. and Camara, P.: A canopy stomatal resistance model for gaseous deposition to vegetated surfaces, Atmos. Environ., 21(1), 91–101, doi:10.1016/0004-6981(87)90274-5, 1987.

Clifton, O. E., Fiore, A. M., Massman, W. J., Baublitz, C. B., Coyle, M., Emberson, L., Fares, S., Farmer, D. K., Gentine, P., Gerosa, G., Guenther, A. B., Helmig, D., Lombardozzi, D. L., Munger, J. W., Patton, E. G., Pusede, S. E., Schwede, D. B., Silva, S. J., Sörgel, M., Steiner, A. L. and Tai, A. P. K.: Dry Deposition of Ozone over Land: Processes, Measurement, and Modeling, Rev. Geophys., 58(1), doi:10.1029/2019rg000670, 2020a.

Clifton, O. E., Paulot, F., Fiore, A. M., Horowitz, L. W., Correa, G., Baublitz, C. B., Fares, S., Goded, I., Goldstein, A. H., Gruening, C., Hogg, A. J., Loubet, B., Mammarella, I., Munger, J. W., Neil, L., Stella, P., Uddling, J., Vesala, T. and Weng, E.: Influence of dynamic ozone dry deposition on ozone pollution, J. Geophys. Res. Atmos., 125(8), e2020JD032398, doi:10.1029/2020jd032398, 2020b.

Emberson, L. D., Kitwiroon, N., Beevers, S., Büker, P. and Cinderby, S.: Scorched earth: How will changes in the strength of the vegetation sink to ozone deposition affect human health and ecosystems?, Atmos. Chem. Phys., 13(14), 6741–6755, doi:10.5194/acp-13-6741-2013, 2013.

Fares, S., Matteucci, G., Scarascia Mugnozza, G., Morani, A., Calfapietra, C., Salvatori, E., Fusaro, L., Manes, F. and Loreto, F.: Testing of models of stomatal ozone fluxes with field measurements in a mixed Mediterranean forest, Atmos. Environ., doi:10.1016/j.atmosenv.2012.11.007, 2013.

Hardacre, C., Wild, O. and Emberson, L.: An evaluation of ozone dry deposition in global scale chemistry climate models, Atmos. Chem. Phys., 15(11), 6419–6436, doi:10.5194/acp-15-6419-2015, 2015.

Hoshika, Y., Osada, Y., de Marco, A., Peñuelas, J. and Paoletti, E.: Global diurnal and nocturnal parameters of stomatal conductance in woody plants and

major crops, Glob. Ecol. Biogeogr., 27(2), 257–275, doi:10.1111/geb.12681, 2018.
Jarvis, P. G.: The Interpretation of the Variations in Leaf Water Potential and Stomatal Conductance Found in Canopies in the Field, Philos. Trans. R. Soc. B Biol. Sci., 273(927), 593–610, doi:10.1098/rstb.1976.0035, 1976.

Lin, M., Malyshev, S., Shevliakova, E., Paulot, F., Horowitz, L. W., Fares, S., Mikkelsen, T. N. and Zhang, L.: Sensitivity of ozone dry deposition to ecosystem-atmosphere interactions: A critical appraisal of observations and simulations, Global Biogeochem. Cycles, 33, doi:10.1029/2018gb006157, 2019.

Mészáros, R., Horváth, L., Weidinger, T., Neftel, A., Nemitz, E., Dämmgen, U., Cellier, P. and Loubet, B.: Measurement and modelling ozone fluxes over a cut and fertilized grassland, Biogeosciences, 6(10), 1987–1999, doi:10.5194/bg-6-1987-2009, 2009. Silva, S. J. and Heald, C. L.: Investigating Dry Deposition of Ozone to Vegetation, J. Geophys. Res. Atmos., 123(1), 559–573, doi:10.1002/2017JD027278, 2018.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyúri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J. P., Valdebenito, A. and Wind, P.: The EMEP MSC-W chemical transport model; Technical description, Atmos. Chem. Phys., 12(16), 7825–7865, doi:10.5194/acp-12-7825-2012, 2012.

Stella, P., Loubet, B., de Berranger, C., Charrier, X., Ceschia, E., Gerosa, G., Finco, A., Lamaud, E., Serça, D., George, C. and Ciuraru, R.: Soil ozone deposition: Dependence of soil resistance to soil texture, Atmos. Environ., doi:10.1016/j.atmosenv.2018.11.036, 2019.

Wang, Y., Jacob, D. J. and Logan, J. A.: Global simulation of tropospheric O 3-NO x-hydrocarbon chemistry - 1. Model formulation, J. Geophys. Res. D Atmos., 103(3339), 10713–10725, doi:10.1029/98jd00158, 1998.

Wesely, M. L.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 41(SUPPL.), 52–63, doi:10.1016/j.atmosenv.2007.10.058, 1989.

Wong, A. Y. H., Geddes, J. A., Tai, A. P. K. and Silva, S. J.: Importance of dry deposition parameterization choice in global simulations of surface ozone, Atmos. Chem. Phys., 19(22), 14365–14385, doi:10.5194/acp-19-14365-2019, 2019.

Zhang, L., Brook, J. R. and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, Atmos. Chem. Phys., 3(6), 2067–2082, doi:10.5194/acp-3-2067-2003, 2003.

2.2 Author's response

Dear referee,
we are thankful for the detailed review. Our replies are below:
In the context of developing an atmospheric (chemical) model we chose to extend the common Wesely scheme of MESSy with well-known empirical relationships. The extension firstly captures the dependency on vegetation density, heat and drought which have been shown to be major drivers of inter-site variability's (Wong et al., 2019, Hardacre et al., 2015). Modelling the stomatal behaviour with more mechanistic models, e.g. based on carbon assimilation is a subject of future developments in MESSy. A paragraph on these future developments will be added as manuscript outlook.
We understand the reviewer doubts when comparing to the dry deposition scheme of other current models. However, the implementations of stomatal conductance dependence on vegetation density, heat and drought stress as well as cuticular uptake linked to meteorology introduce firstly important functionalities of dry deposition at vegetation to MESSy. Although the scheme is still only based on four different surface types these revision represents a significant advancement for dry deposition modelling with MESSy allowing a more realistic account of an important global ozone sink. Thereby, MESSy still lacks a detailed and mechanistic description of terrestrial vegetation that is evaluated and routinely used by the MESSy community. The documentation, evaluation and publication of the developments presented in the manuscript are important beyond the MESSy community. In fact EMAC participates to the world wide Model Intercomparison Projects (not at least CMIP6), where the full documentation of the models published is essential to understand differences among the different models. To provide a platform for this kind of model description is one of the goals of GMD. Implementing a biome-dependent dry deposition model coupled to CO2 assimilation (White et al. 2004) is planned as a follow-up development in MESSy. Biome-dependent vegetation cover information, required for this scheme, are then provided by global input data which, however, represent only the annual cycle of vegetation. The recently available dynamic vegetation model LPJ-GUESS providing detailed vegetation information with the temporal variability required for a climate model could be a further improvement. By now the one-way coupling of LPJ-GUESS as a MESSy submodel is only in the initial evaluation of the coupling with the atmospheric model (Forrest et al., 2020). A description of these future developments will be added as an outlook section to the manuscript.
We are aware of the limitations of the Jarvis-type model but among others Fares et al. (2013) showed that the Jarvis-type model captured measured O3 dry deposition fluxes better than a Ball-Berry model based on CO2 assimilation. The criticism of the Jarvis-type model in Fares et al. (2013) concerns the missing ability of the VPD factor in representing the 'VPD driven afternoon depression'. However, we used instead of the proposed drought stress factor by Jarvis the mechanistic factor based on the optimised exchange of CO2 and water by plants (Katul et al. 2009). We will add a section on the uncertainties and limitations

of the Jarvis-type model to the manuscript.
We see the importance of the biome-dependent parameters which however can introduce uncertainties since they are assigned to measurements whereas the absolute values are influenced by multiple factors like genotype and local climatic conditions (Sulis et at., 2015; Hoshika et al., 2018, Tuovinen et al., 2009). Admittedly, detailed parameters are presented in e.g. LRTAP (2009) but for large-scale models with their limitations they have to be simplified like it is done for the EMEP model (Simpson et al. 2012). The most sensitive and uncertain parameter for dry deposition modelling at stomata g_{smax} is not used. Instead, we parametrized the background stomatal behaviour explicitly depending on the photosynthetically active radiation according to Sellers (1985). Regarding the optimal temperature of stomatal behaviour we have to consider that for the maximum and minimum temperature, which are directly related to the optimal temperature, only less measurements under field conditions are available (Hoshika et al., 2018). For these reasons among others, we decided to keep the four-type surface scheme of MESSy for dry deposition modelling in which then biome-dependent parameter sets are not included.
Yes, the discrepancy at ATTO could be due to an improper parametrization of stomatal conductance whereas the neglected chemical within-canopy reactions, however, are also an uncertainty source (Freire et al. 2017). On the other hand the biased meteorology and moisture cycling is a well-known issue in ECHAM (Hagemann and Stacke 2015) and plays a role for dry deposition modelling here as well. In Fig. 5b of the manuscript we can show that modified meteorology and transpiration at least partly improves the modelled dry deposition velocity in the Amazon forest.
The cuticular parametrization by Zhang et al. (2002) was implemented in order to account for the second important ozone deposition pathway in our model. This pathway was effectively neglected in the previous model version. As well as for the stomatal uptake we built up on the existing resistance scheme in MESSy which distinguish between only four different surface types. Here we also used less generalised parameters. An overall consideration of the uncertainty and limitations of the used model, however, is important and will be added as a separate section to the manuscript.
The whole data comparison at the four chosen sites account for the most important high vegetation covered biomes on the Earth. For the reason of uniqueness and importance to investigate atmospheric processes in a remote and pristine forest the Amazonian Tall Tower Observatory (ATTO) stands out. In order to include an analysis at this site in our study we adapt the choice of the simulation period to the availability of measurements there, specifically. The used and described measurement data listed in e.g. Clifton et al. (2020a) have been obtained in the late 2000s and early 2010s. However, the analysis period should cover the recent decade which includes most extreme drought and heat events (where the stomatal stress factors are aimed for). Moreover, since we consider the inter-annual differences at the different locations we only compare data which cover the same time period. Including further measurement sites would require a new simulations.

The changes, indeed affect trace gases other than ozone. However, this manuscript focuses on ozone because among it's atmospheric importance the applied Wesely scheme is based on the the dry deposition mechanism of ozone (Wesely 1989). By including the changes in O_x budget, that includes NO_2 and HNO_3, we cover many important tropospheric trace gases. We will further add a figure with the changes for the fluxes of NO_2, HNO_3, HCHO and SO_2 and the respective description.
Indeed, the linear scaling lead to an overestimation of the uptake. Thank you for pointing to this typo.
We can add discussions and acknowledgements on existing soil deposition parametrizations.
The wet skin fraction is calculated from the wet skin reservoir (wl [m]) and Leaf Area Index (LAI $[m^2/m^2]$:

$$\sim wl/(1 + LAI) \tag{1}$$

whereas the snow covered fraction depends mainly on the snow at the surface (h_s [m water equivalent]):

$$cvs \sim \tanh{(h_s)}\sqrt{h_s} \tag{2}$$

The detailed description can be found in the documentation of ECHAM5 (Klimarechenzentrum 1992 eq. 3.3.2.4; Roeckner et al., 2003 eq. 6.45)
We reviewed and ask for several data sets. The chosen data sets were the best available of ozone dry deposition (flux data and ozone mixing ratio or velocity data) with the required temporal resolution and coverage which also represent different parts and biomes of the world. As examples, for Harvard forest data of O3 dry deposition flux and O3 mixing ratio is only available until 1997[1]) whereas at Blodgett forest the total measuring period (2001-2007) doesn't match the chosen simulation period. Like described above we didn't use data with non-matching time coverage since we consider inter-annual differences at the measurement sides.
Yes, at the points where the uptake to the leaf surfaces is meant the term cuticular conductance should be used. However, some cited studies report measurements (partioning) of non-stomatal dry deposition which captures among others the removal at the cuticle. We will clearly distinguish this terms.

References

Forrest, Matthew, et al. "Including vegetation dynamics in an atmospheric chemistry-enabled general circulation model: linking LPJ-GUESS (v4. 0) with the EMAC modelling system (v2. 53)." Geoscientific Model Development 13.3 (2020).

[1] data coverage of 'O3.mlb' and f.o3 is shown in plot 3 and 5 in https://harvardforest1.fas.harvard.edu/sites/harvardforest.fas.harvard.edu/files/data/plots/hf004-01.pdf

Freire, L. S., et al. "Turbulent mixing and removal of ozone within an Amazon rainforest canopy." Journal of Geophysical Research: Atmospheres 122.5 (2017): 2791-2811.

Hagemann, Stefan, and Tobias Stacke. "Impact of the soil hydrology scheme on simulated soil moisture memory." Climate Dynamics 44.7-8 (2015): 1731-1750.

Hardacre, C., O. Wild, and L. Emberson. "An evaluation of ozone dry deposition in global scale chemistry climate models." Atmospheric Chemistry and Physics 15.11 (2015): 6419-6436.

LRTAP: "Mapping critical levels for vegetation, in: Manual on Methodologies and Criteria for Mapping CriticalLoads and Levels and Air Pollution Effects, Risks and Trends." Revision of 2009, edited by Mills, G., UNECE Convention on Long-range Transboundary Air Pollution. International Cooperative Programme on Effects of AirPollution on Natural Vegetation and Crops

Klimarechenzentrum, Deutsches. "The ECHAM3 atmospheric general circulation model." Techn. Rep 6 (1992).

Hoshika, Yasutomo, et al. "Global diurnal and nocturnal parameters of stomatal conductance in woody plants and major crops." Global ecology and biogeography 27.2 (2018): 257-275.

Roeckner, Erich, et al. "The atmospheric general circulation model ECHAM 5. PART I: Model description." (2003).

Sellers, Piers J. "Canopy reflectance, photosynthesis and transpiration." International journal of remote sensing 6.8 (1985): 1335-1372. Wong, Anthony Y., et al. "Importance of dry deposition parameterization choice in global simulations of surface ozone." Atmospheric Chemistry and Physics 19.22 (2019).

Sulis, M.; Langensiepen, M.; Shrestha, P.; Schickling, A.; Simmer, C. & Kollet, S. J. Evaluating the Influence of Plant-Specific Physiological Parameterizations on the Partitioning of Land Surface Energy Fluxes J. Hydrometeor, American Meteorological Society, 2015, 16, 517-533

Tuovinen, Juha-Pekka, Lisa Emberson, and David Simpson. "Modelling ozone fluxes to forests for risk assessment: status and prospects." Annals of Forest Science 66.4 (2009): 1-14.

Wesely, Ml. "Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models." Atmospheric environment 23.6 (1989): 1293-1304.

Zhang, Leiming, Jeffrey R. Brook, and Robert Vet. "On ozone dry deposition—with emphasis on non-stomatal uptake and wet canopies." Atmospheric Environment 36.30 (2002): 4787-4799.
White, P. W. "IFS documentation CY23r4: Part IV physical processes." (2004).

2.3 Author's changes to the manuscript

- Line 34, 74, 77, 213: 'non-stomatal' was deleted; line 56,58,175: 'non-stomatal' was replaced by 'cuticular'
- Section 9 as an outlook was added
- In Section 2.2 (line 133-144)

> *Due to the importance of stomatal and cuticular uptake for ozone dry deposition these pathways are considered in this study. The parametrisations and revisions are described in the following sections.An investigation of the soil resistance might also be desirable (Schwede et al., 2011; Fares et al., 2012) but the proposed parametrisations are not well established yet and therefore not studied here*

was replaced and extended with

> *Due to the importance of stomatal and cuticular uptake for ozone dry deposition their respective parametrisations are modified in this study (see Sec. 2.2.1 and 2.2.2). Also, ozone deposition to soil might be an important pathway (Schwede et al., 2011; Fares et al., 2012) but process understanding remains limited due to scant observational constraints (Clifton et. al., 2020a, b). Stella et al.(2011) showed an exponential increase of soil resistance with surface relative humidity in three agricultural datasets which, however, varies much between different sites (Stella et al., 2019) and contradicts with previous findings (Altimir et al., 2006; Lamaud et al., 2002; Zhang et al., 2002). Models by e.g. Mezaros et al. (2009) and Lamaud et al. (2009) apply a linear dependence on soil water content for parameterising soil resistance. These parametrisations rely on input variables like the minimum soil resistance (Stella et al., 2011) which introduce an uncertainty due to measurement constraints. Also, the performance of a mechanistic model as proposed by Clifton et al. (2020b) depends on many input variables and parameters whose estimation is challenging and mostly biome-dependent. Due to these uncertainties and limitations the current parametrization of soil resistance in MESSy (see Kerkweg et al. (2006) for details) was not modified in this study.*

- In Section 4 (line 272-277), the following were added

> *The chosen data sets are the best available of ozone dry deposition (flux data and ozone mixing ratio or velocity data) with the required temporal resolution and coverage of diverse biomes of the world. The*

analysis is aimed at covering the recent decade which includes the most extreme drought and heat events (where the stomatal stress factors are aimed for). For the reason of uniqueness and importance of atmospheric processes in a remote and pristine forest like the Amazon Basin we included measurements from there among others the Amazonian Tall Tower Observatory (ATTO).

- Line 129: 'underestimates' was changed to 'overestimates'
- In Section 5 (line 416-433):

> *In total, the changes by the revised dry deposition scheme increase the multiyear mean (2010-2015) loss by dry deposition from 946 Tg/yr to 1001 Tg/yr for ozone and from 978 Tg/yr to 1032 Tg/yr for odd oxygen ([...]) which is in the reported range (Hu et al., 2017; Young et al., 2018). Accordingly, (boreal) summer ground-level ozone over land is reduced by up to 12 ppb (24%) peaking over Scandinavia, Asia, central Africa and East Canada (Fig. 6d). In the Northern Hemisphere, also the zonal mean of the tropospheric ozone mixing ratio show a noticeable reduction far from the ground compared to the default scheme (Fig. 9c). This has the potential to reduce the positive bias of tropospheric ozone on the Northern Hemisphere (20 %) reported by Young et al. (2018). The reduction of ground-level ozone due to the change in dry deposition is a combined effect of the impact on ozone deposition and the altered loss of soluble oxygenated VOCs which are ozone precursors at ground level. The change of dry deposition decreases HCHO at ground level (Fig. 9b) by up to 25 % in (boreal) summer*

were replaced by

> *In total, the changes by the revised dry deposition scheme increase the multiyear mean (2010-2015) loss of ozone by dry deposition from 946 Tg/yr to 1001 Tg/yr (Hu et al., 2017; Young et al., 2018). Accordingly, (boreal) summer ground-level ozone over land is reduced by up to 12 ppb (24%) peaking over Scandinavia, Asia, central Africa and East Canada (Fig. 6d). In the Northern Hemisphere, also the zonal mean of the tropospheric ozone mixing ratio show a noticeable reduction far from the ground compared to the default scheme (Fig. 9c). This has the potential to reduce the positive bias of tropospheric ozone on the Northern Hemisphere (20 %) reported by Young et al. (2018). However, besides ozone also other atmospheric tracer gases are affected by the change in dry deposition. The global annual dry deposition flux of odd oxygen ([...]), which includes many important troposheric trace gases, increases from Tg/yr to 1032 Tg/yr due to the revision. This is in good agreement with the reported numbers by Hu et al. (2017) and Young et al. (2018). In Fig. 9, we show additionally the absolute and relative change of the multi-year annual*

average dry deposition loss of SO_2, NO_2,HNO_3 and HCHO. As a very soluble species the loss of SO_2 is increased by the revised dry deposition scheme whereas the predefined low cuticular and wet skin resistance of HNO_3 in the old scheme were replaced with the new mechanism leading to an decrease in dry deposition. The altered loss of NO_2 and HCHO and other ozone precursors at ground level, especially soluble oxygenated VOCs contributes to the total change in ozone loss. NO_2 is deposited almost 40 % more significantly contributing to the net reduction in ozone production but is mostly counterbalanced by other processes. The change of HCHO dry deposition flux is small on a global and annual scale and only important regionally, most in (boreal) summer, when it decreases HCHO at ground level (Fig. 9b) by up to 25 %.

3 Comment 3

3.1 Comment from referree

General comments:
This manuscript presents a revised dry deposition scheme in EMAC model. Revision includes improved stomatal deposition dependence on vegetation density, two meteorological factors and soil moisture, and improved non-stomatal deposition dependence on meteorological and environmental factors. Then authors evaluated the impact of this revision on ozone dry deposition on daily and seasonal scales and explored the effect of each separate parameterization. In general, I think this manuscript is really well written, and it meets the criteria for publication on Geophysical Model Development:
- the manuscript contributes to the modeling of boundary layer dynamics and atmospheric chemistry
- scientific approach and methods used are valid, results are discussed in an appropriate way
- simulation is reproducible because of data and code availability, and detailed description in the main manuscript and supplements

Just a few questions and details need to be further addressed before accepting for publication:

Specific comments:

- Abstract could be revised to better summarize (shorten) the model development details and leave room for model performance improvement and impact on global ozone budget, etc.

- Authors selected 6 land types out of 11 in the model. Could authors add in the reason for including or excluding certain land types?

- Mismatching meteorology: in Sect. 4.2, authors choose/have to use meteorology data from ERA5 to assess the impact of stress factors on the diurnal cycle of dry deposition. And in line 371-372, '. . . , as the humidity over the Amazon forest **is probably too low** in the model'. Same argument is presented in line 415-417. Could these mismatches/comparisons in meteorology be shown in appendix as figures?

Technical corrections:

- Line 92, 'The the'.
- Line 286, is 'what' a typo here?

3.2 Author's response

Dear referee, many thanks for the comments. We appreciate that the referee recognise the purpose and the importance of this manuscript. Please find our replies to the special comments below. We agree that the abstract can be more concise to include additional important aspects you mentioned. We will re-write it.
We used the already existing surface scheme in MESSy for this study. The 6 land types (better termed surface types) is a generalisation of the originally given 11 types ((1) Urban land, (2) agricultural land, (3) range land, (4) deciduous forest, (5) coniferous forest, (6) mixed forest including wetland, (7) water including both salt and fresh, (8) barren land - mostly desert, (9) non-forested wetland, (10) mixed agricultural and range land, (11) rocky open areas with low-growing shrubs) whereas e.g. the here used surface type vegetation represents all vegetated areas.
Yes, the mentioned aspect plays an important role for the analysis at ATTO. The argumentation can be illustrated and clarified with figures of the comparison of the meteorology. We will add these figures to the appendix.

The typos mentioned in the technical comments will be corrected.

3.3 Author's changes in the manuscript

- In the abstract (Line 9-14):

> *.The default dry deposition scheme has been extended with adjustment factors to predict stomatal responses to temperature and vapour pressure deficit. Furthermore, an explicit formulation of the non-stomatal deposition to the leaf surface (cuticle) dependent on humidity has been implemented based on established schemes. Finally, the soil moisture availability function for plants has been revised to be consistent with the simple hydrological model available in EMAC. This revision was necessary in order to avoid unrealistic stomatal closure where the model shows a strong soil dry bias, e.g. in the Amazon basin in the dry season.*

were replaced by

> *including meteorological adjustment factors for stomatal closure and an explicit cuticular pathway.*

We added additionally (line 17/18):

> *The scheme is limited by a small number of different surface types and generalised parameters.*

and line 24/25:

> *The change of ozone dry deposition is also reasoned by the altered loss of ozone precursors.*

- In Section 2.2 (line 105/106) we added:

> *This was adapted by Ganzeveld and Lelieveld (1995) to the surface scheme of the ECHAM climate model(Klimarechenzentrum et al., 1992).*

- Doubling 'the' (line 114) was removed
- Line 326: 'what' was changed to 'which'
- Figure A3 on the difference of temperature and relative humidity between EMAC and ERA5 at ATTO was newly included in the appendix (reference in measurement and gl. impact section)

A revised dry deposition scheme for land-atmosphere exchange of trace gases in ECHAM/MESSy v2.54

Tamara Emmerichs[1], Astrid Kerkweg[1], Huug Ouwersloot[2], Silvano Fares[3], Ivan Mammarella[4], and Domenico Taraborrelli[1]

[1]Institute of Energy and Climate Research 8, Troposphere, Forschungszentrum Jülich, Jülich, Germany
[2]Max Planck Institute for Chemistry, Mainz, Germany
[3]National Research Council, Institute of Bioeconomy, Rome, Italy
[4]Institute for Atmospheric and Earth System Research / Physics, Faculty of Science, University of Helsinki, Finland

Correspondence: Domenico Taraborrelli (d.taraborrelli@fz-juelich.de)

Abstract. Dry deposition to vegetation is a major sink of ground-level ozone and is responsible for about 20 % of the total tropospheric ozone loss. Its parametrisation in atmospheric chemistry models represent a significant source of uncertainty for the global tropospheric ozone budget and might account for the mismatch with observations. The model used in this study, the Modular Earth Submodel System (MESSy2) linked to ECHAM5 as an atmospheric circulation model (EMAC), is no exception. Like many global models, EMAC employs a “resistance in series” scheme with the major surface deposition via plant stomata which is hardly sensitive to meteorology, depending only on solar radiation. Unlike many global models, however, EMAC uses a simplified high resistance for non-stomatal deposition which makes this pathway negligible in the model. However, several studies have shown this process to be comparable in magnitude to the stomatal uptake, especially during the night over moist surfaces. Hence, we present here a revised dry deposition in EMAC . The default dry deposition scheme has been extended with adjustment factors to predict stomatal responses to temperature and vapour pressure deficit. Furthermore, an explicit formulation of the non-stomatal deposition to the leaf surface (cuticle) dependent on humidity has been implemented based on established schemes. Finally, the soil moisture availability function for plants has been revised to be consistent with the simple hydrological model available in EMAC. This revision was necessary in order to avoid unrealistic stomatal closure where the model shows a strong soil dry bias, e.g. in the Amazon basin in the dry seasonincluding meteorological adjustment factors for stomatal closure and an explicit cuticular pathway. These modifications for the three stomatal stress functions have been included in the newly developed MESSy submodel VERTEX, i.e. a process model describing the vertical exchange in the atmospheric boundary layer, which will be evaluated for the first time here. The scheme is limited by a small number of different surface types and generalised parameters. The MESSy submodel describing the dry deposition of trace gases and aerosols (DDEP) has been revised accordingly. The comparison of the simulation results with measurement data at four sites shows that the new scheme enables a more realistic representation of dry deposition. However, the representation is strongly limited by the local meteorology. In total, the changes increase the dry deposition velocity of ozone up to a factor of 2 globally, whereby the highest impact arises from the inclusion of cuticular uptake, especially over moist surfaces. This corresponds to a 6 % increase of global annual dry deposition loss of ozone resulting globally in a slight decrease of ground-level ozone but a regional decrease of up to 25 %. The change of ozone dry deposition is also reasoned by the altered loss of ozone precursors.

Thus, the revision of the process parameterisation as documented here has among others the potential to significantly reduce the overestimation of tropospheric ozone in global models.

1 Introduction

Ground-level ozone is a secondary air pollutant which is harmful for humans and ecosystems. Besides chemical destruction, a large fraction of it is removed by dry deposition which accounts for about 20 % of the total O_3 loss (Young et al., 2018). The process description of dry deposition considers boundary-layer meteorology (e.g. turbulence), chemical properties of the trace gases and surface types. In most global models, dry deposition of trace gases is parameterised using the "resistance in series" analogy by Wesely (1989). The largest deposition rates of ozone occur over dense vegetation (Hardacre et al., 2015) where it mainly follows two pathways, through leaf openings (stomata) and to leaf waxes (cuticle~~, non-stomatal~~) (Fares et al., 2012). Thereby, stomatal uptake is commonly parameterised following the empirical multiplicative approach by Jarvis (1976) which uses a predefined minimum resistance and multiple environmental response factors like in Zhang et al. (2003); Simpson et al. (2012); Emberson et al. (2000).

More advanced formulations often used by land surface models (Ran et al., 2017; Val Martin et al., 2014) are based on the CO_2 assimilation by plants during photosynthesis (Ball et al., 1987; Collatz et al., 1992). Both approaches rely on the choice and constraints of ecosystem dependent parameters and have different advantages (Lu, 2018). A further role in coupling stomata to ecosystems play stomatal optimization models whereas optimal stomatal activity with a maximum amount of carbon gain and a minimum loss of water is calculated based on eco-physiological processes (e.g., Cowan and Farquhar, 1977) . Of particular interest are stomatal optimization models which, based on eco-physiological processes, maximize carbon gain while minimizing water loss. According to Wang et al. (2020) these models are promising in representing stomatal behaviour and improving carbon cycle modelling. Non-stomatal deposition has been less investigated by now therefore most models use predefined constant resistances or scale it with leaf area index (e.g., Val Martin et al., 2014; Simpson et al., 2012) while some apply an explicit parametrization based on the observational findings of enhance cuticular uptake under leaf surface wetness (Altimir et al., 2006).

The different parametrisations of the (surface) resistances cause main model uncertainties in computing dry deposition fluxes of trace gases, which depend on the response to hydroclimate and land-type specific properties (Hardacre et al., 2015; Wu et al., 2018; Wesely and Hicks, 2000). ~~A model intercomparison by Schwede et al. (2011), however, points to the parametrisation of soil and cuticular uptake as source of uncertainty. For instance, Val Martin et al. (2014) found that the reported positive ozone bias (10-20 % Northern Hemisphere) can be attributed to an oversimplification of the dry deposition scheme. Also, Wong et al. (2019) has shown that discrepancies of up to 8 ppb difference in ground-level ozone arise from different parametrisations. The~~ Thereby, it has been shown that the original Wesely-based parametrisation generally captures well the seasonal and diurnal cycle of dry deposition velocity whereas model-observation discrepancy at seasonal scales arises from biased land type and leaf area index input data (Silva and Heald, 2018). Wong et al. (2019) stated that discrepancies of up to 8 ppb in ground-level ozone arise from different parametrisations.

The current dry deposition scheme of EMAC uses 6 surface types ~~(original: 11 and 5 seasonal categories) whereas the parametrised processes are for the~~ where the parameterised processes represent the forest canopy as a whole (big-leaf approach). Thereby, the uptake over vegetation relies on stomatal deposition as the only pathway determined by the photosynthetically active radiation (Kerkweg et al., 2006). According to Fares et al. (2012) and Rannik et al. (2012) the stomatal uptake in parametrisations often lacks the dependence on meteorological and environmental variables (leaf area index, temperature, vapour pressure deficit). Moreover, several studies (e.g., Hogg et al., 2007; Fares et al., 2012; Clifton et al., 2017) found the contribution of an additional process to dry deposition at the leaf covering of plants. Zhang et al. (2002) firstly derived a parametrisation from field studies which establishes the important link of this process to meteorology. ~~Furthermore~~In general, findings by Solberg et al. (2008); Andersson and Engardt (2010); Wong et al. (2019) highlight the importance of considering the dry deposition-meteorology dependence in global models. Such an extension would realistically enhance the sensitivity of dry deposition to climate variability and would result in a more accurate prediction of ground-level ozone.
Given the importance of ozone as a major tropospheric oxidant, air pollutant and greenhouse gas, an accurate representation of dry deposition is desirable (Jacob and Winner, 2009). Additionally, the significance of a realistic representation of land-atmosphere feedbacks rises in the light of the changing Earth's climate with projected increase of extreme events frequency and intensity (Coumou and Rahmstorf, 2012).

Here, we present a revision of the existing Wesely's based dry deposition scheme ~~, incorporating recent~~ in MESSy which has a very simplified representation of vegetation and soil. The modifications are done by well-established findings about the controls of stomatal and ~~non-stomatal uptake~~ cuticular uptake of trace gases. The calculation of stomatal deposition fluxes is extended by including the vegetation density, two meteorological adjustment factors and an improved soil moisture availability function for plant stomata following the multiplicative algorithm by Jarvis (1976). For the first time in MESSy, a parametrisation for ~~non-stomatal~~ cuticular dry deposition dependent on important meteorological and environmental variables is implemented explicitly (Zhang et al., 2003). In Sect. 2, a description of the model set up and the simulations is provided whereas especially the transition to the new vertical exchange scheme is described in detail. Subsequently, the new scheme VERTEX is evaluated. In Sect. 4, the impact of the changes on ozone dry deposition is evaluated on daily and seasonal scales by comparison with measurements at four different sites. Here, advantages, uncertainties and missing processes in the revised scheme are identified. Next, the global impact on ground-level ozone is assessed by separating the effect of the different implemented parametrisations. Then, Sect. 6 provides a description of the uncertainties in modelling stomatal conductance and Sect. 7 comprises an investigation of the sensitivity to model resolution. Sect. 8 ~~, finally,~~ summarises the main findings and the remaining process and model uncertainties ~~. These~~ which form the basis for the provided recommendations.

Sect.9 describes future planned developements.

2 Model description

This study uses the Atmospheric Chemistry Model ECHAM/MESSy. The Modular Earth Submodel System (MESSy v2.54) (Jöckel et al., 2010) provides a flexible infrastructure for coupling processes to build comprehensive Earth System Models

(ESMs) and is utilised here with the fifth generation European Centre Hamburg general circulation model (Roeckner et al. (2003), ECHAM5) as atmospheric general circulation model. The dry deposition process of gases is calculated within the submodel DDEP (Kerkweg et al., 2006). This is described in Section 2.2. It relies on the vertical exchange submodel VERTEX (Sect. 2.1), former E5VDIFF, which contains the calculation of stomatal uptake (Eq. 5) and soil moisture stress (Eq. 12). The stomatal uptake parametrisation is the base for the evapotranspiration scheme in VERTEX (Appendix B) which also incorporates the soil moisture stress.

2.1 The new vertical exchange submodel VERTEX

The submodel VERTEX represents land-atmosphere exchange and vertical diffusion as an alternative to the default submodel E5VDIFF in ECHAM5/MESSy. In 2016 Huug Ouwersloot branched VERTEX off from E5VDIFF. He optimised the code and applied bug fixes. This includes changes in calculation of the transfer coefficients for vertical diffusion, the latent heat vaporisation, the convective transfer coefficient, the storage of the friction velocity, the roughness length over sea, the kinematic heat and moisture fluxes and the 2 m and 10 m friction velocity. A detailed description can be found in the Supplement.

2.2 Dry deposition over vegetation

Dry deposition of trace gases to vegetation is calculated according to the multiple resistance scheme by Wesely (1989) shown in Figure 1. The scheme, originally designed for a regional model with 11 land types and 5 seasonal categories, is used here with 6 generalized land types (Kerkweg et al., 2006). This was adapted by Ganzeveld and Lelieveld (1995) to the surface scheme of the ECHAM climate model (Klimarechenzentrum et al., 1992). The vegetation canopy is represented as one system , i.e., the detailed structure and plant characteristics are neglected (one big-leaf approach). Only one assumption about the canopy structure is made: the leaves are horizontally oriented and the leaf density is uniformly vertically distributed (Sellers, 1985). This is required in the formula for the calculation of stomatal resistance (Eq. 5).

The resistances (in $\mathrm{s\,m^{-1}}$) in the big-leaf approach account for mass and energy transfer mainly exerted by the boundary layer turbulence (R_a), molecular diffusion via the quasi-laminar boundary layer (R_{qbr}) and heterogeneous losses at the surface (R_s) (Kerkweg et al., 2006). With these, the dry deposition velocity v_d of a trace gas X (in $\mathrm{s\,m^{-1}}$) is defined as follows:

$$v_d(X) = \frac{1}{R_a + R_{qbr}(X) + R_s(X)} \quad (1)$$

The ~~the~~ dry deposition flux $f_d(X)$ (in $\mathrm{molecules\,m^{-2}\,s^{-1}}$) is determined by multiplying the dry deposition velocity with the trace gas concentration $C(X)$ (in $\mathrm{molecules\,m^{-3}}$):

$$f_d(X) = -v_d(X) \cdot C(X) \quad (2)$$

The total resistance over land combines the resistances over snow, soil, vegetation (veg) and wet skin (ws) weighted by the respective land covered fraction of a grid box (Kerkweg et al., 2006). In the following, only the latter two are considered. The resistances R_a and R_{qbr} are commonly parameterised with standard formulations from micro-meteorology (Kerkweg et al.,

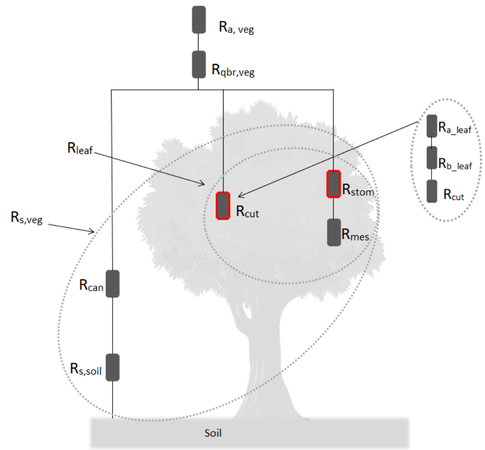


Figure 1. Dry deposition resistance analogy, modified resistors are marked with red boxes (adapted from Zhang et al. (2003)).

2006; Wesely and Hicks, 1977). For the surface resistance over vegetation ($R_{s,veg}$) the parametrisation according to Zhang et al. (2003) is used:

$$\frac{1}{R_{s,veg}(X)} = \frac{1}{R_{can} + R_{s,soil}(X)} + \underbrace{\frac{1}{R_{cut}(X)} + \frac{1}{R_{stom,corr}(X) + R_{mes}(X)}}_{R_{leaf}(X)} \tag{3}$$

which consists of the soil resistance ($R_{s,soil}(X)$), the in-canopy aerodynamic resistance (R_{can}) (as in Kerkweg et al. (2006)) and the leaf resistance ($R_{leaf}(X)$). The gas uptake by leaves ($leaf$) can be separated in two parallel pathways: the cuticular (cut) and the stomatal ($stom$) with its associated mesophyllic pathway (mes), where the latter has negligible resistance for ozone and highly soluble species (Wesely, 1989). In contrast to the default formulation in MESSy (Eq. A1), the resistances in the updated scheme are provided at canopy scale in order to avoid linear scaling with the Leaf Area Index (LAI, area of leaves [m^2]/surface area [m^2]). In fact, the linear scaling of resistances with LAI assumes that the leaves act in parallel and ~~underestimates~~ overestimates the uptake for high LAI values (>3-4) (Ganzeveld et al., 1998; Baldocchi et al., 1987). Furthermore, the quasi-laminar boundary resistance of individual leaves is included through the cuticular deposition scheme (see Sect. 2.2.2) whereas $R_{qbr,veg}$ is a separate term in the old formulation (Eq. A1).

Due to the importance of stomatal and cuticular uptake for ozone dry deposition ~~these pathways are considered~~ their respective parametrisations are modified in this study ~~. The parametrisations and revisions are described in the following sections.An investigation of the soil resistance might also be desirable (Schwede et al., 2011; Fares et al., 2012) but the proposed parametrisations are not well established yet and therefore not studied here~~(see Sec. 2.2.1 and Sec. 2.2.2). Also, ozone deposition to soil might be an important pathway (Schwede et al., 2011; Fares et al., 2012) but process understanding remains limited due to scant observational constraints (Clifton et al., 2020b, a). Stella et al. (2011) showed an exponential increase of soil resistance with surface relative humidity in three agricultural datasets which, however, varies much between different sites (Stella et al., 2019) and contradicts with previous findings (Altimir et al., 2006; Lamaud et al., 2002; Zhang et al., 2002). Models by e.g. Mészáros et al. (2009); Lamaud et al. (2009) apply a linear dependence on soil water content for parameterising soil resistance. These parametrisations rely on input variables like the minimum soil resistance (Stella et al., 2011) which introduce an uncertainty due to measurement constraints. Also, the performance of a mechanistic model as proposed by Clifton et al. (2020b) depends on many input variables and parameters whose estimation is challenging and mostly biome-dependent. Due to these uncertainties and limitations the current parametrization of soil resistance in MESSy (see Kerkweg et al. (2006) for details) was not modified in this study.

2.2.1 Uptake through plant stomata

The stomata are actively regulated openings between the plant cells. They are scattered mostly over the lower (hypostomatous) epidermis of leaves. They control the H_2O and CO_2 exchange by plants which is the essential coupling of vegetation to the atmosphere and therefore to weather and climate. Here, the default parametrisation of stomatal resistance (Eq. A2) is extended by adding dependencies on meteorological variables according to the Simple Biosphere Model (SiB) by Sellers et al. (1986) based on previous work by Jarvis (1976) for temperature (T) and vapour pressure deficit (VPD):

$$R_{stom,corr}(X) = \frac{R_{stom}(PAR,LAI)}{f(W_s)\cdot f(T)\cdot f(VPD)}\cdot\frac{D_{H_2O}}{D(X)} \tag{4}$$

The optimal stomatal resistance for water ($R_{stom}(PAR,LAI)$) is corrected with the ratio of the molecular diffusivity of the species ($D(X)$) and water (D_{H_20}). The optimal stomatal resistance depends on the photosynthetically active radiation (PAR) and Leaf Area Index (LAI) (Ganzeveld and Lelieveld, 1995; Sellers, 1985):

$$R_{stom}(PAR,LAI) = \frac{kc}{\left[\frac{b}{dPAR} ln\left(\frac{d\exp(kLAI)+1}{d+1}\right) - ln\left(\frac{d+\exp(-kLAI)}{d+1}\right)\right]} \tag{5}$$

where $\mathrm{k} = 0.9$ is the extinction coefficient, $\mathrm{c} = 100\ \mathrm{s\,m^{-1}}$ is the minimum stomatal resistance and $a = 5000\ \mathrm{J\,m^{-3}}$, $b = 10\ \mathrm{W\,m^{-2}}$ and $d = \frac{a+b\cdot c}{c\cdot PAR}$ are fitting parameters (Sellers, 1985). For historical reasons, LAI was set to 1 in order to obtain the stomatal resistance at leaf level (Ganzeveld and Lelieveld, 1995). This has been changed and the seasonal evolution of stomatal resistance now follows the LAI which, in our study, is based on a 5-year climatology of monthly Normalised Differential Vegetation Index (NDVI) satellite data (Ganzeveld et al., 2002).

First, the stomatal resistance is corrected by the inverse of the temperature stress factor ($1/f(T)$) derived by Jarvis (1976):

$$f(T) = b_3(T-T_l)(T_h-T)^{b_4}$$

$$f(T) = b_3(T-T_l)(T_h-T)^{b_4} \tag{6}$$

$$b_3 = (T_0-T_l)(T_h-T_0)^{-b_4} \tag{7}$$

$$b_4 = (T_h-T_0)/(T_h-T_l) \tag{8}$$

where the empirical parameters are T_h =318.15 K, T_l =268.15 K ~~, b_3 =8and b_4=0.5~~and T_0 =298.15 K.

Secondly, following the analysis by Katul et al. (2009), a stress factor dependent on vapour pressure deficit $(1/f(VPD))$ was added to the calculation of stomatal resistance in VERTEX:

$$p_{H_2O,sat}(T) = 0.61078 \exp\left(\frac{17.1 \cdot T(p_{H_2O})}{235 + T(p_{H_2O})}\right) \quad (9)$$

$$VPD = p_{H_2O,sat}(T) - p_{H_2O} = \left(1 - \frac{RH}{100}\right) p_{H_2O,sat}(T) \quad (10)$$

$$f(VPD) = VPD^{-\frac{1}{2}} \quad (11)$$

with $T(p_{H_2O})$ (in K) as the surface temperature, p_{H_2O} (in kPa) as the pressure of water vapour and $p_{H_2O}(T)$ [kPa] the pressure of saturated air. The vapour pressure deficit is calculated according to Kraus (2007).

While the stomatal resistance at canopy scale is actually calculated within the MESSy submodel VERTEX, the submodel DDEP uses it for the calculation of dry deposition fluxes. Thus, in DDEP the user can choose between the old scheme based on Ganzeveld and Lelieveld (1995) and the new scheme actually using the stomatal resistance at canopy scale. The latter is activated by setting the DDEP $\&CTRL$ namelist parameter $l\_ganzeori$ to $.FALSE.$. How the stomatal resistance is calculated is chosen in VERTEX by the $\&CTRL$ namelist parameter $irstom$.

- $irstom = 0$ activates the original parametrisation.
- Separate modifications:
  - $irstom = 2$: variable LAI,
  - $irstom = 3$: T dependency,
  - $irstom = 4$: VPD dependency, respectively.
- $irstom = 5$: all modifications.
- $irstom = 1$: stomatal resistance with variable LAI at leaf scale. Instead of choosing LAI=1 in Eq. 5 to represent the stomatal resistance at leaf level, as is done by the original code, Eq. (5) is calculated at canopy level using the actual LAI and then multiplied by LAI to obtain the average stomatal resistance at leaf level. For this case, the DDEP namelist parameter $l\_ganzeori$ have to be set to .TRUE.

The stomatal activity of plants and the strength of surface-atmosphere coupling strongly depend on the parameterised plant-water stress (Combe et al., 2016). The soil water budget is represented by a "bucket scheme" where the soil water in a single layer is prescribed by a geographically varying predefined field capacity and soil wetness governed by transpiration, precipitation, runoff, snow melt and drainage (Roeckner et al., 2003). This scheme is used by so called "first-generation" models. However, EMAC controls evapotranspiration through the stomatal resistance (Appendix B) which is the most important feature of biophysical ("second-generation") land-surface models. Thereby, the stomatal resistance is calculated often like the one described here (Eq. 4) including temperature, VPD and soil moisture stress (Seneviratne et al., 2010; Sellers et al., 1997). The originally used plant-water stress function of Jarvis (1976) and Sellers et al. (1986), however, relies on leaf water potential ($f(\psi)$) for different plant types, which is difficult to estimate. Hence, EMAC uses a plant-water stress function dependent on soil moisture ($f(W_s)$. The default parametrisation (Eq. A, $ifws = 0$ in VERTEX $\&CTRL$) applies as lower threshold the permanent wilting point of plants (W_{pwp}, 35% of field capacity[1]) in the calculation of the soil moisture stress factor ($f(W_s)$). However, soil moisture is significantly underpredicted by the model in some regions and the calculated $f(W_s)$ can be 0 for long periods. This is unrealistic and effectively shuts down dry deposition, e.g. during the dry season in the Amazon region. For this reason $f(W_s)$ is parameterised here according to the original formulation by Delworth and Manabe (1988) removing the lower limit:

$$f(W_s) = \begin{cases} 1 & W_s(t) > W_{cr} \\ \frac{W_s(t)}{W_{cr}} & W_s(t) \leq W_{cr} \end{cases} \tag{12}$$

where $W_s(t)$ is the surface soil wetness (in m). W_{cr} (in m) is defined as the critical soil moisture level (75 % of the field capacity) at which the transpiration of plants is reduced. The modified parametrisation in Eq. 12 can be applied by setting the $\&CTRL$ parameter $ifws = 1$ in the VERTEX namelist.

2.2.2 Cuticular deposition

According to several field studies (e.g., Van Pul and Jacobs, 1994; Hogg et al., 2007; Fares et al., 2012) ~~non-stomatal~~ cuticular deposition is an important contributor to ozone uptake and should not be neglected in models. Therefore, an explicit parametrisation of cuticular deposition as used in many North American air quality modelling studies (Huang et al., 2016; Kharol et al., 2018) has been implemented. The ~~non-stomatal~~ gas uptake by leaf surfaces is based on two parallel routes, for which an analogy to ozone (highly reactive) and sulphur dioxide (very soluble) is used. The cuticular resistance is calculated as:

$$R_{cut}(X) = \frac{R_{cut,d}(\mathrm{O_3})}{10^{-5} \cdot H(X) + s_{reac}(X)} \tag{13}$$

where $H(X)$ is the effective Henry's law coefficient as measure for the solubility. The reactivity of a species is rated by the parameter s_{reac}. For highly reactive species ($s_{reac} = 1$) the same property as for ozone is assumed (second term in Eq. 13), while for less reactive species ($s_{reac} = 0.1, 0$) the uptake is effectively reduced (Wesely, 1989). For soluble species, the uptake at wet skin is assumed to be similar to the one of sulphur dioxide and is calculated as:

$$R_{ws}(X) = \left[\frac{1/3}{R_{cut,w}(SO_2)} + 10^{-7} \cdot H(X) + \frac{s_{reac}(X)}{R_{cut,w}(\mathrm{O_3})} \right]^{-1} \tag{14}$$

where $R_{cut,w}(\mathrm{SO_2})$ and $R_{cut,w}(\mathrm{O_3})$ are the resistances of sulphur dioxide and ozone at wet surfaces, respectively. The constant values of the default formulae (Eq. A4, A5) are replaced by parametrisations which account for the meteorological dependence

[1] maximum amount of water the soil can hold against gravity over periods of several days

of cuticular uptake according to Zhang et al. (2002):

$$R_{cut,d}(\mathrm{O_3/SO_2}) = \frac{R_{cut,d0}(\mathrm{O_3/SO_2})}{\exp(0.03 \cdot RH) \cdot LAI^{0.25} \cdot u_*} \tag{15}$$

$$R_{cut,w}(\mathrm{O_3/SO_2}) = \frac{R_{cut,w0}(\mathrm{O_3/SO_2})}{LAI^{0.5} \cdot u_*} \tag{16}$$

where the cuticular resistance of $\mathrm{O_3}$ and $\mathrm{SO_2}$, respectively, is distinguished for dry canopies ($R_{cut,d}$) and wet canopies ($R_{cut,w}$) depending on relative humidity (RH in $\%$), Leaf Area Index (LAI in $\mathrm{m^2\,m^{-2}}$) and friction velocity (u_* in $\mathrm{m\,s^{-1}}$). The input parameters are $R_{cut,d0}$($\mathrm{O_3}$)=5000 $\mathrm{s\,m^{-1}}$, $R_{cut,w0}$($\mathrm{O_3}$)=300 $\mathrm{s\,m^{-1}}$ and $R_{cut,d0}$($\mathrm{SO_2}$)=2000 $\mathrm{s\,m^{-1}}$ (Zhang et al., 2002). For rain and dew conditions, values of 50 $\mathrm{s\,m^{-1}}$ and 100 $\mathrm{s\,m^{-1}}$ are prescribed for $R_{cut,w0}(SO_2)$. In contrast to traditional approaches, these parametrisations also consider the aerodynamic and the quasi-laminar boundary resistances of individual leaves. For the usage in MESSy this can be switched on via $l\_ganzeori = .FALSE.$ in the $\&CTRL$ namelist of DDEP.

2.3 Simulations

In order to answer the different research questions of this study, two different types of simulations have been performed (Tab. 1):

(1) Simulations to investigate dry deposition and the effect of the modifications in VERTEX:

These simulations are based on the Chemistry-Climate Model Initiative (CCMI) setup (Jöckel et al., 2016). To allow for comparison with measurements, the model dynamics have been nudged towards realistic meteorology by the assimilation of data from the European Centre for Medium-range Weather Forecasting (ECMWF) (Jöckel et al., 2010). Additionally, the QCTM mode is used, i.e., the chemistry does not feed back to the dynamics, resulting in the same meteorology for all simulations (Deckert et al., 2011). All modifications for the dry deposition scheme are employed in a 7-year simulation (REV, 2009-2015). Additionally, a 1.5-year simulation covering the period 2017 to July 2018 (2017 as spin-up) has been performed to cover the measurement periods (Sect. 4). For the same periods simulations with the same configuration except applying the default dry deposition scheme (DEF) have been conducted. The individual effects of the different modifications are investigated by two 2-year simulations employing the different namelist switches (Sect. 2.2). Moreover, a free-running sensitivity simulation with an additional temperature and drought stress factor for evapotranspiration (Appendix B) has been performed aiming at an improved representation of local meteorology especially in the Amazon. The station simulation output and the global output are analysed in Sections 4 and 5, respectively. In addition, two 2-year simulations are realised for different horizontal resolutions (REST42, REST63) to investigate the resolution dependency of dry deposition (Sect. 7). All these simulations use 31 model layers with the top at 10 hPa and take the first year of simulation as spin-off.

(2) Simulations for the evaluation of VERTEX as boundary layer scheme:

Two pure dynamical (i.e., without chemistry) 30-year simulations with the old (clim-E5) and the new boundary layer description (clim-VER), respectively, have been performed.

All simulations were performed at the Jülich Supercomputing Center with the JURECA Cluster (Jülich Supercomputing Centre, 2018).

Table 1. List of EMAC simulations

Simulation	Spatial resolution	Time period	Remarks
(1) **Dry deposition mechanism: CCMI chemistry, nudged, no feedbacks (QCTM)**			
REST42	T42L31 (2.8° x2.8°)	2009/2010	irstom=5, ifws=1, l_ganzeori=F
REST63	T63L31 (1.9° x1.9°)	2009/2010	irstom=5, ifws=1, l_ganzeori=F
REV (revised)	T106L31 (1.1° x1.1°)	2009-2015, 2017-June 2018	irstom=5, ifws=1, l_ganzeori=F
DEF (default)	T106L31 (1.1° x1.1°)	2009-2015, 2017-June 2018	default ddep scheme
REV-fws	T106L31 (1.1° x1.1°)	2009/2010	irstom=5, ifws=0, l_ganzeori=F
REV-fTfD	T106L31 (1.1° x1.1°)	2009/2010	irstom=2, ifws=1, l_ganzeori=F
REV-NNTR	T106L31 (1.1° x1.1°)	2014/2015	free-running, all ddep modifications (as REV), all stress factors applied to evapotranspiration (izwet=1).
(2) Climatology comparison: no chemistry, free-running			
clim-E5	T42L90 (2.8° x2.8°, up to 0.01 hPa)	1979-2008	E5VDIFF for vertical exchange
clim-VER	T42L90 (2.8° x2.8°, up to 0.01 hPa)	1979-2008	VERTEX for vertical exchange

3 VERTEX evaluation

In order to advise the usage of VERTEX (with the default settings) as the default vertical exchange submodel in MESSy the dynamics produced by both submodels are compared. Therefore, two dynamical, free running, 30-year simulations have been performed using the E5VDIFF or the VERTEX submodels, respectively. To obtain a comparable radiative imbalance at TOA (top of the atmosphere) with VERTEX the four cloud parameters have been tuned in advance according to Mauritsen et al. (2012). The tuning factors can be found in Table 2. The radiative imbalance at TOA is slightly positive at present-day conditions (Mauritsen et al., 2012; Stephens et al., 2012), here E5VDIFF gives a negative value. The difference between the tuned VERTEX and E5VDIFF is small and within the uncertainty range of $\pm 0.4\ \mathrm{W\,m^{-2}}$.

Additionally, global mean values of surface temperature, cloud liquid water, relative humidity and planetary boundary layer height of EMAC using E5VDIFF and EMAC using VERTEX with the respective uncertainty range for the period 1979-2008 are represented in Figure 2. The results for cloud liquid water and planetary boundary height show no significant differences between the VERTEX and E5VDIFF simulation since each annual means falls in the confidence interval of the other. This is not always the case for surface temperature and relative humidity. However, the 30-year means of surface temperature and relative humidity simulated by E5VDIFF and VERTEX are not significantly different.

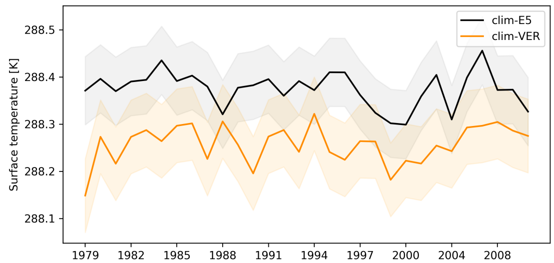
Table 2. Overview of tuning parameter settings and global mean properties

Parameters		**EMAC(E5VDIFF)**	**EMAC(VERTEX)**
Cloud mass-flux above level of non-buoyancy		0.3	0.3
Entrainment rate for shallow convection		$1e-3$	$1e-3$
Entrainment rate for deep convection		$1e-4$	$1e-4$
Conversion rate to rain in convective clouds		$1.5e-4$	$1.6e-4$
Properties	**Observed** [a]	**EMAC(E5VDIFF)**	**EMAC(VERTEX)**
Total cloud cover [%]		67.12	67.27
Water vapour path [$\mathrm{kg\,m^{-2}}$]		25.03	24.83
Liquid water path [$\mathrm{kg\,m^{-2}}$]		0.077	0.077
Total precipitation [mm/d]		1.28	1.31
Surface net shortwave [$\mathrm{W\,m^{-2}}$]	152-167	158.27	158.32
Surface net longwave [$\mathrm{W\,m^{-2}}$]	-(40-57)	-54.82	-54.93
Surface sensible heat flux [$\mathrm{W\,m^{-2}}$]	-(16-19)	-18.75	-19.65
Surface latent heat flux [$\mathrm{W\,m^{-2}}$]	-(75-87)	-87.45	-88.73
Planetary albedo [%]		32.38	32.37
Shortwave net at TOA [$\mathrm{W\,m^{-2}}$]	238-244	230.99	231.00
Longwave net at TOA [$\mathrm{W\,m^{-2}}$]	-(237-241)	-232.46	-232.55
Radiation imbalance at TOA [$\mathrm{W\,m^{-2}}$]		-1.47	-1.55

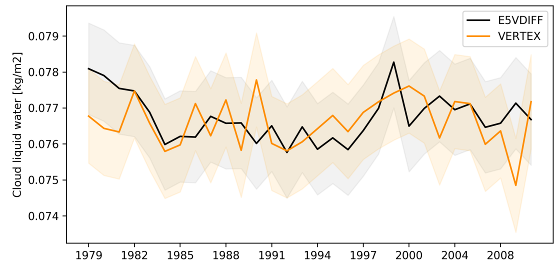
[a] Stevens and Schwartz (2012)

4 Evaluation with deposition measurements

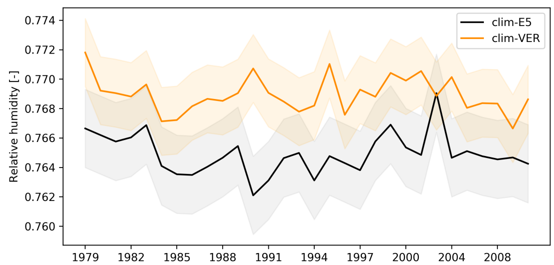
To assess the impact of the code revision/modifications on the variability of dry deposition we compare the sensitivity simulations DEF, REV, REV-fTfVPD, REV-fws and REV-NNTR (see Tab. 1, all at T106L31 resolution) with dry deposition measurements at four field sites (listed in Table 3). The chosen data sets are the best available of ozone dry deposition (flux data and ozone mixing ratio or velocity data) with the required temporal resolution and coverage of diverse biomes of the world. The analysis is aimed at covering the recent decade which includes the most extreme drought and heat events (where the stomatal stress factors are aimed for). For the reason of uniqueness and importance of atmospheric processes in a remote and pristine forest like the Amazon Basin we included measurements from there among others the Amazonian Tall Tower Observatory (ATTO). Ozone dry deposition fluxes were measured with the eddy covariance and gradient method (Ontario). From this data, deposition velocities were calculated by the means of ozone concentration data. The eddy covariance technique determines a turbulent flux by the covariance of the measured vertical velocity and the gas concentration. Due to the stochastic nature of turbulence, these measurements have an uncertainty of 10 to 20 % under typical observation conditions (Rannik et al., 2016). For the gradient method used at Borden forest research station the dry deposition flux was estimated from con-



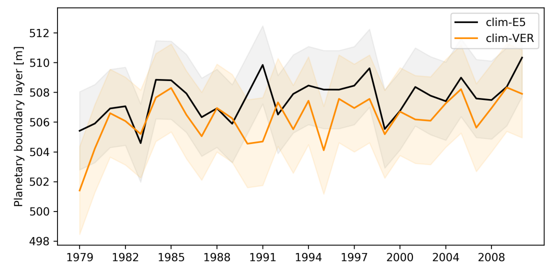
(a) Surface temperature



(b) Vertical integrated cloud liquid water



(c) Relative humidity



(d) Planetary boundary layer height

Figure 2. Global mean properties and the uncertainty range (95. confidence interval in shaded) of the climatology simulations with E5VDIFF (clim-E5) and with VERTEX (clim-VER) for the period 1979-2008.

centration gradients below and above the canopy and the eddy diffusivity according to the Monin-Obukhov similarity theory. The estimated dry deposition velocities (V_d) show an uncertainty of $\approx 20\ \%$ which is due to the assigned canopy, the inherent limitations of the algorithm and the measurement uncertainties in concentrations. However, results are in good agreement with other eddy covariance measurements (Wu et al., 2016).

4.1 Annual cycle of dry deposition

The annual cycle of dry deposition is mainly driven by the evolution of vegetation and is generally represented well in models (Silva and Heald, 2018). We use here the long time series measured at Borden and Hyytiälä to identify the impact of the code modifications on the annual cycle of dry deposition velocity. The available micro-meteorological data help to distinguish the different effects. From the hourly data, we calculated multiyear (2010-2012) monthly means. To explore the contribution of stomatal and cuticular uptake, the individual velocities are calculated for O_3 according to the model calculations (Kerkweg

Table 3. Dry deposition measurements. In the description of vegetation/climate the reported Leaf Area Index (LAI, in $m^2\,m^{-2}$) is given in brackets, v_d^{mod} and v_d^{obs} are the average measured and modelled dry deposition velocity.

Site	Vegetation/climate	Location (height)	Time period	$v_d^{mod}(v_d^{obs})$ $cm\,s^{-1}$	Reference
Hyytiälä, Southern Finland (SMEARII)	boreal forest, Scots Pine, (LAI=3-4)/ cold temperate	61.85N 24.28E (22 m/16 m[a])	2010-2012	0.29 (0.28)	Keronen et al. (2003)
Lindcove research station, California (US)	Citrus Orchard (LAI=3)/ Mediterranean	36.35N 119.09W (131 m)	Oct.2009-Nov.2010	0.22 (0.49)	Fares et al. (2012) and Fares[b]
Borden research station, Ontario, Canada	mixed forest (LAI=4.6)/ temperate	44.19N 79.56W (33 m)	2010-2012	0.34 (0.47)	Wu et al. (2018)
Amazonian Tall Tower (ATTO), Manaus, Brazil	rainforest (LAI=6)/ tropical humid	2.15S -59.01W (41 m)	November 2015, April/May 2018	0.18 (0.67), 0.33 (1,0)	available on request: Matthias Sörgel (m.soergel@mpic.de)

[a]Meteorological measurement height
[b]Ozone data is not available here

et al., 2006):

$$G_{cut,d} = \frac{(1-ws)\cdot(1-cvs)\cdot veg}{R_{cut,d}(\mathrm{O_3})} \qquad G_{cut,w} = \frac{ws\cdot(1-cvs)}{R_{cut,w}(\mathrm{O_3})} \tag{17}$$

$$G_{ns} = G_{cut,d} + G_{cut,w} \tag{18}$$

$$G_{stom} = \frac{(1-ws)\cdot(1-cvs)\cdot veg}{R_{stom,corr}(\mathrm{O_3})} \tag{19}$$

$$v_p = \frac{G_p}{G_{stom}+G_{ns}}\cdot v_d \tag{20}$$

where G names the individual conductances (inverse of resistance) of stomata ($stom$), dry cuticle (cut,d), wet cuticle (cut,w) and non-stomata (ns). veg, ws and cvs give the vegetation fraction, the wet skin fraction and the snow covered fraction, respectively. G_p and v_p are the individual conductance and the velocity of one pathway. Further terms are described in Sect. 2.2. The multiyear (2010-2012) annual cycle of the simulated dry deposition velocity at Borden forest (Fig. 3a) captures the observed cycle well until June. The new scheme reproduces the observations better than the old scheme. This is a consequence of the increase in nighttime mean velocities due to the much larger cuticular contribution (Fig. A1a, A1b). However, due to the overestimated stomatal uptake in the default scheme (see Sect. 2.2.1) only slight deviations from the new dry deposition scheme are visible in the daily mean shown in Figure 3a. The mismatch of the simulated and measured V_d from August to

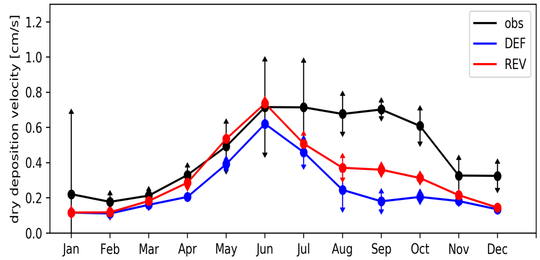
October is a consequence of the underestimation of relative humidity leading to too low simulated cuticular deposition (Fig. 3c, 3e). This effect exceeds the impact of the overestimation of relative humidity (only) in summer, because the LAI is higher in summer. In general, the cuticular uptake parametrisation accounting for LAI, friction velocity, RH and surface wetness conditions performs, in our simulations, better than parametrisations without these dependencies as expected from the study of Wu et al. (2018). Unfortunately, the cuticular uptake parametrisation also introduces uncertainties to the modelled non-stomatal uptake. Moreover, accounting for biogenic volatile organic compounds (BVOCs) like in Makar et al. (2017) would enhance in-canopy loss of ozone, significantly increase non-stomatal dry deposition and lead to improved simulation results (Wu et al., 2018). The representation of in-canopy air chemistry is outside the scope of the present study but planned within a subsequent study.

In contrast, the amplitude of the annual cycle and the mean of dry deposition fluxes in Hyytiälä are overestimated by both schemes during spring and summer (Fig. 3b). For the default scheme, this is due to the oversimplification of the stomatal uptake that only accounts for a constant LAI of 1 $\mathrm{m^2\,m^{-2}}$ (see Sect. 2.2.1) which is far from the measured LAI of 3-4 $\mathrm{m^2\,m^{-2}}$ during this period (Keronen et al., 2003). Enabling the new scheme (REV), increases the dry deposition velocity which reproduces the measured values in autumn better. The contribution of non-stomatal dry deposition of 25-45 $\%$ during day reported by Rannik et al. (2012) is represented partly by that. However, the new scheme leads to an even higher overestimation by the model from April to July. The sensitivity simulation REV-fws (default $f(W_s)$) points to the increase of soil moisture stress function (see Sect. 2.2.1, Eq. 12) as one reason for the overestimation of V_d in summer (Fig. 3b, A2b). Moreover, the overestimation in June/July is partly ($\sim$ 10 $\%$) due to the too high model LAI compared to the measured values of 3-4 (Fig. A2a). The remaining gap (Fig. 3f) can be explained by restricting the analysis to wet conditions (RH $>$70 $\%$) only, and the analysis of the sensitivity simulation REF-fTfD (no $f(T)$ and $f(VPD)$). This suggests that the overestimated V_d (Fig. A2c) in summer is due to the stress factors for stomatal uptake since the modelled and measured temperature mismatch. VPD has been identified by Rannik et al. (2012) as a strong driver of day-time total deposition velocity ~~what~~ which confirms the importance of inclusion of VPD dependence for stomatal uptake.

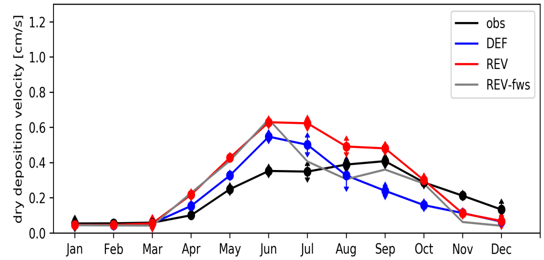
4.2 Importance of stress factors for the diurnal variation of deposition

The short-term measurements at Lindcove research station and at Amazonian Tall Tower Observatory (ATTO) are used to assess the impact of the stress factors on the diurnal cycle of dry deposition velocity in spring and summer. Additionally, micrometeorological and additional flux data make possible to consider the stomatal resistance ($\sim$inverse of the velocity, calculations according to Fares et al. (2012)) and the underlying meteorological conditions. Since the respective micro-meteorological measurements are not available at ATTO, data extracted from the ERA5 global climate reanalysis at the 1000 hPa pressure level (Copernicus, 2017) is used here.

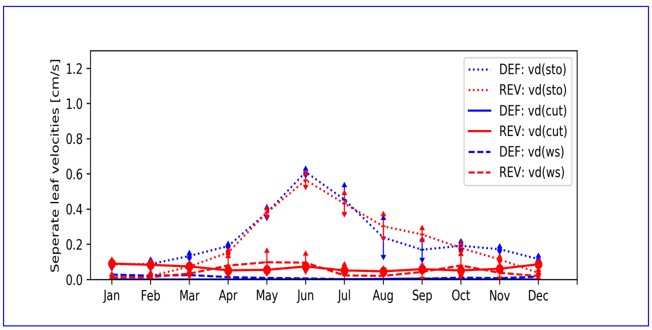
The diurnal cycle of dry deposition velocity at the Lindcove research station follows the solar variation (Fares et al., 2012) and is generally well reproduced by the model with the best match in spring (Fig. ~~4~~4a). The revised dry deposition scheme reduces the underestimation of measured night-time V_d due to the inclusion of cuticular uptake, which Fares et al. (2012) identified as an important ozone sink for exactly this measurement site. The measured dry deposition velocity increases at sunrise (around



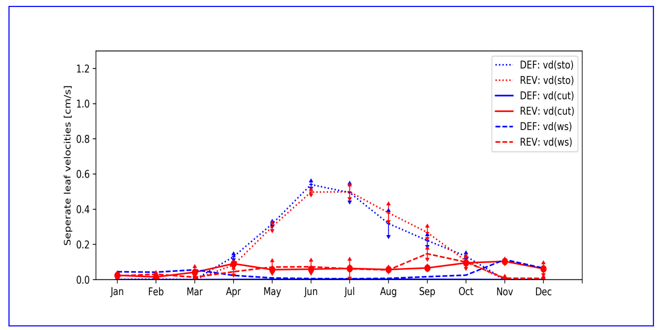
(a) Dry deposition velocity at Borden forest



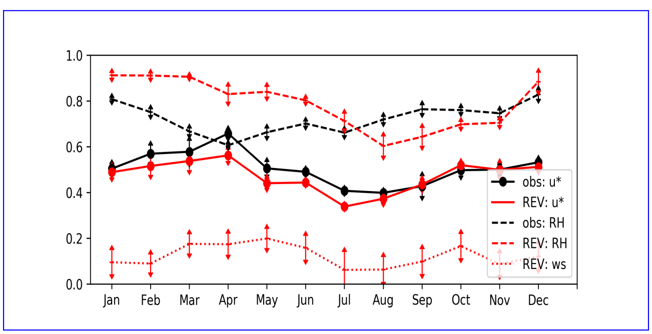
(b) Dry deposition velocity at Hyytiälä



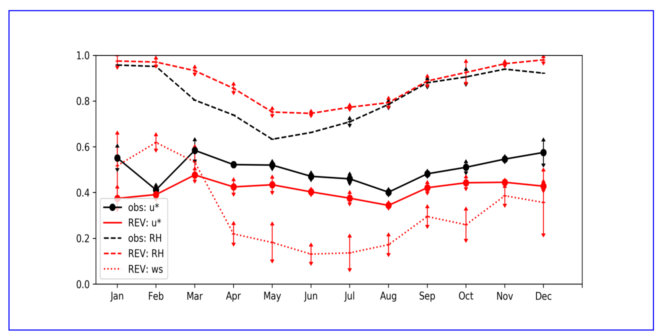
(c) Contributing deposition velocity (vd) from the model



(d) Contributing deposition velocities (vd) from the model



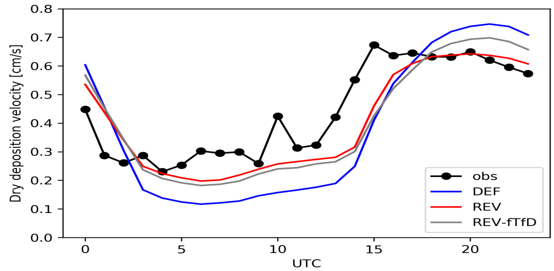
(e) Friction velocity (u*), Relative humidity (RH), Wet skin fraction (ws)



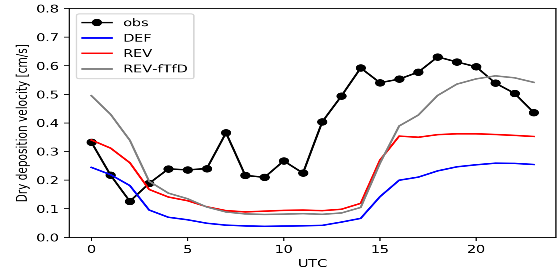
(f) Friction velocity (u*), Relative humidity (RH), Wet skin fraction (ws)

Figure 3. Measured (obs) and modelled (DEF, REV) multiyear mean (2010-2012) and REV-fws (2010) annual cycle. Left: Borden forest, right: Hyytiälä, for (a) and (b) arrows give 1σ

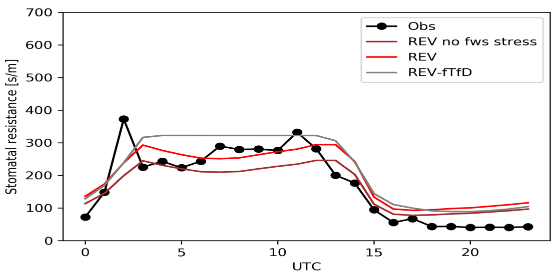
15 UTC) and remains almost constant during the day. This is only reproduced by the revised dry deposition scheme. The comparison of the dry deposition velocity from the revised scheme (red line) and the velocity without stomatal T and VPD stress (gray line) in Figure 4a illustrates the necessity of accounting for the stress factors. This is consistent with Fares et al. (2012) who report a high negative correlation of $V_d(sto)$ with VPD and temperature and relates it to stomatal stress. The direct comparison of the stomatal resistances calculated from measured and modelled variables (Fig. 4c) shows an improvement of the



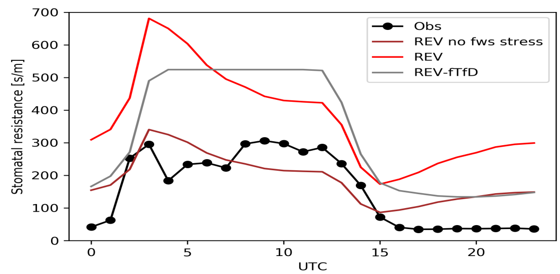
(a) Dry deposition velocity in spring 2010



(b) Dry deposition velocity in summer 2010



(c) Stomatal resistance in spring 2010



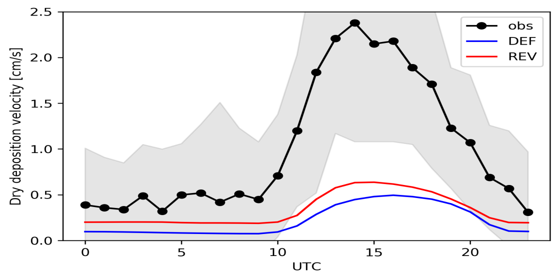
(d) Stomatal resistance in summer 2010

Figure 4. Diurnal cycles of measured (obs) and modelled (DEF,REV,REV-fTfD) ozone dry deposition velocity and stomatal resistance in spring and summer 2010 at Lindcove research station.

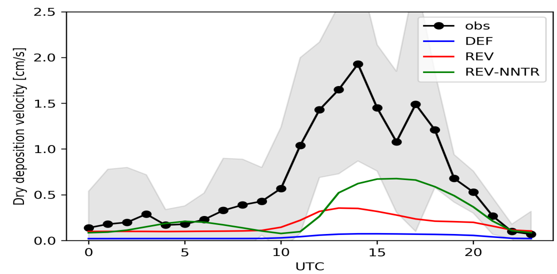
modelled resistances (comparing DEF an REV). However, the modelled daytime stomatal resistance is still too high compared to the measurements. This points to an underestimation of stomatal uptake by the model during day. A small fraction can be explained by the direct effect of the stomatal soil moisture stress in the model which does not occur in reality since the Citrus Orchard was watered during the measurement campaign. Contrastingly in summer, the model underestimation of V_d is higher than in spring (Fig. 4b). As seen from the comparison of stomatal resistance values (Fig. 4d) the model underestimates the stomatal uptake. This is because the irrigation of the Orchard leads to cooling sustained evapotranspiration and keeps $f(T)$ low. Thus in the model, a too high temperature stress act on the stomata. Moreover, neglecting the soil moisture stress on stomata would bring the stomatal resistance values closer since the irrigation at the site ensures a constant and high soil moisture. The irrigation of the Citrus Orchard during day also enhances surface wetness and favours deposition at cuticles (Fares et al., 2012; Altimir et al., 2006) which cannot be captured by the model. Fares et al. (2012) estimate the stomatal contribution to only account for 20-45 $\%$ of the total daytime dry deposition flux during both seasons and point to soil deposition and reactions of ozone with NO and VOCs as major sinks at Citrus Orchard, especially during flowering season. The contribution of these pathways is expected to be enhanced by the inclusion of further biogenic VOCs within the chemical mechanism and the explicit parametrisation of in-canopy residence and transport.

Tropical forests are known to be effective O_3 sinks with observed mean midday maximum dry deposition velocity of 2.3 $\mathrm{cm\,s^{-1}}$ (Rummel et al., 2007) due to much higher LAI compared to other sites (e.g. Lindcove). The measured dry deposition velocity at ATTO shown in Fig. 5a and Fig. 5b is no exception but shows a high variability (standard deviation). The diurnal cycle follows the solar radiation with maximum V_d at 15 UTC and highest amplitude during the wet season (April/May 2018). The amplitude of the diurnal cycle is highly underestimated in both EMAC simulations with the highest mismatch during daytime. This is similar for other models. In fact, Hardacre et al. (2015) report a general and large underestimation of dry deposition velocities by models over tropical forests with highest predicted values of 0.25 $\mathrm{cm\,s^{-1}}$. Here, the simulation with the revised dry deposition scheme (REV) shows only a minor increase of V_d during the wet season. Since stomatal uptake is known to be an important daytime sink (Freire et al., 2017), the underestimation of the total dry deposition flux is partly attributed to a too low simulated stomatal uptake caused by the overestimation of temperature and the underestimation of relative humidity (Fig. A3). The increase of dry deposition velocity by the new scheme is mainly due to the lowered soil moisture stress on stomata ($f(W_s)$) shown in Fig. 5e. Freire et al. (2017) also links stomatal uptake to the efficiency of turbulent mixing in transporting ozone down to the canopy. In general, 10 $\%$ of the total ozone sink during daytime and 39 $\%$ during night is associated with in-canopy processes (Freire et al., 2017). Freire et al. (2017) and Bourtsoukidis et al. (2018) identified the oxidation of sesquiterpenes as an important contributor to the chemical nighttime sink. Cuticular deposition might also play a role in humid conditions during night (Rummel et al., 2007) which is underestimated by the model due to the biased relative humidity (Fig. 5c).

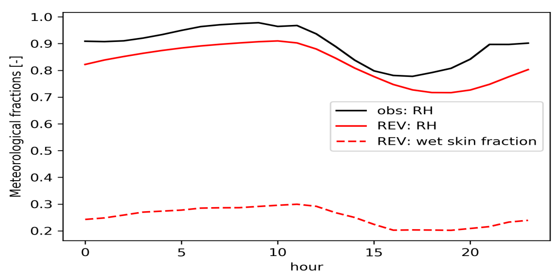
The uncertainty introduced by the mismatching meteorology becomes even more obvious when comparing measurements and simulations for November 2015. This month was characterised by temperatures of 2 to 3 °C above average and unusual little rainfall (compared to usual conditions in this season) due to a strong El Nino event (National Centers for Environmental Information). The dryness is overestimated by the model with a too high temperature (Δ=+5 to +8 K), too low relative humidity (Δ=-30 to -40 $\%$)) and too dry soil. The lack of available soil moisture ($f(W_s)$=0) effectively shuts down stomatal deposition in the default simulation (DEF), whereas the modification of the soil moisture stress function (neglecting the artificial lower limit, see Eq. 12) in the revised model (REV) allows for an increased deposition (Fig. 5b). The temperature and relative humidity biases result in corresponding mismatching stress factors for the stomata that are double the ones derived from reanalysis data (Fig. 5f). This mismatch leads to an underestimation of stomatal uptake. This result is confirmed by the sensitivity simulation REV-NNTR for which no meteorological nudging has been applied and the stress factors $f(T)$ and $f(VPD)$ are also used for the calculation of evapotranspiration. The REV-NNTR simulation yields much more realistic results compared to the measurements capturing at least 50 $\%$ of the measured V_d during day (Fig. 5b). This improvement is partly due to the omission of nudging. As the latter can have a detrimental effect on precipitation and evaporation (Jeuken et al., 1996). The temperature bias of the model is associated with the missing soil moisture buffer simulated by the bucket scheme. Incorporating a 5-layer scheme has been shown to lead to a more realistic soil water storage capacity especially in the Amazon and to a removal of this bias (Hagemann and Stacke, 2013). Nevertheless, the REV-NNTR simulation suggests that the stress factors $f(T)$ and $f(VPD)$ significantly contribute to buffer soil moisture and ameliorate the dryness bias.



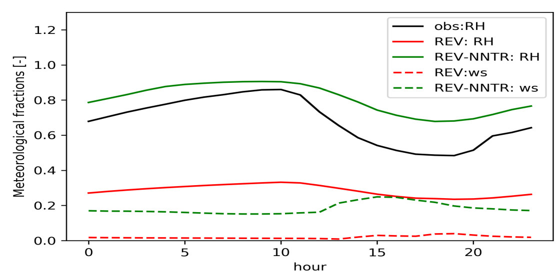
(a) Dry deposition velocity in April-May 2018



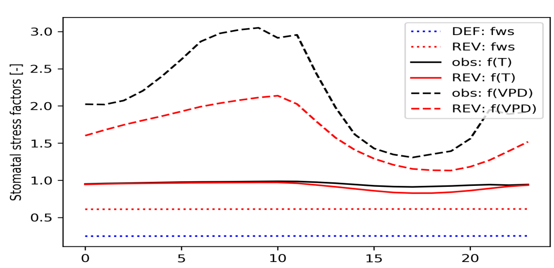
(b) Dry deposition velocity in November 2015



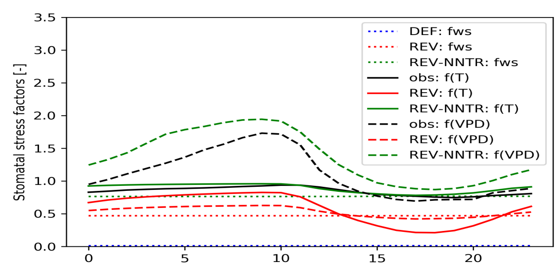
(c) Relative humidity (RH) and wet skin fraction (ws) in April-May 2018



(d) Relative humidity (RH) and wet skin fraction (ws) in November 2015



(e) Stomatal stress factors in April-May 2018



(f) Stomatal stress factors in November 2015

Figure 5. Diurnal cycles of measured (obs) and modelled (DEF, REV, REV-NNTR: free-running $f(T)$ and $f(VPD)$ for evapotranspiration) ozone dry deposition velocities in wet and dry season at ATTO (gray: standard deviation).

5 Global impact on ground-level ozone

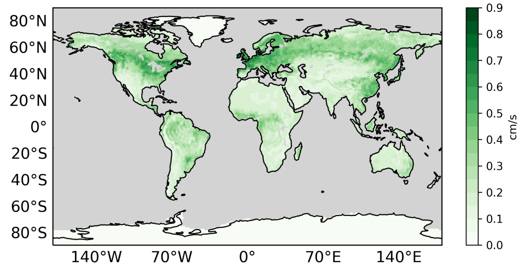
Given the importance of dry deposition for ground-level ozone and the uncertainty of dry deposition parametrisations in models (Young et al., 2018; Hardacre et al., 2015) the global impact of the implemented code changes is assessed in this section. The global (boreal) summer mean distributions of deposition velocity and ground-level mixing ratio for O_3 shown in Figs. 6a/6b are generally in the same range as reported for global models (e.g. Val Martin et al. (2014); Hardacre et al. (2015)). However, like most global models, EMAC overestimates tropospheric ozone in comparison to satellite observations (Righi et al., 2015). Applying the revised dry deposition scheme increases the mean summer V_d by up to 0.5 $\mathrm{cm\,s^{-1}}$ (Fig. 6c). The highest fraction of this increase arises from the inclusion of cuticular uptake at wet surfaces ($V_{cut,w}$) (Fig. A4b). The effect is large over the most northern continental regions (Fig. 6f) and even more pronounced where LAI is high like in Scandinavia and East Canada (for LAI distribution see Fig. A4a). Additionally, the uptake at dry surfaces ($V_{cut,d}$) is enhanced with up to 0.3 $\mathrm{cm\,s^{-1}}$ higher dry deposition velocity (Fig. 6e). This is because the default scheme applies a very high constant resistance for this process.

Concerning the stomatal deposition, the impacts of three different stress factors are considered. First, over relatively dry soil, i.e., where soil moisture exceeds 35 % of field capacity (wilting point of plants), the soil moisture stress is reduced by the modified parametrisation. Neglecting the plants' wilting point as the lower limit for soil moisture stress on stomata weakens the dependency on field capacity. Thus, dry deposition is enhanced by up to ~~0.25~~0.32 $\mathrm{cm\,s^{-1}}$ as illustrated in Figure 7a. Second, the inclusion of temperature and (third) VPD adjustment factors, indeed, leads to a spatially varying impact of $\pm$ 0.27 $\mathrm{cm\,s^{-1}}$ change in V_d (Fig. 7b). In humid and cold temperate regions, like Siberia and Canada, no temperature stress appears and the VPD adjustment factor increases the stomatal uptake. In East U.S., Kazakhstan and Central Amazon during boreal summer stomata are stressed by temperature and VPD. This effect is overpredicted by the model, as the humidity over the Amazon forest is probably too low in the model (see Fig. A3). The stress factors are shown in Figure ~~A4c and A4d .~~ A4d and A4c.

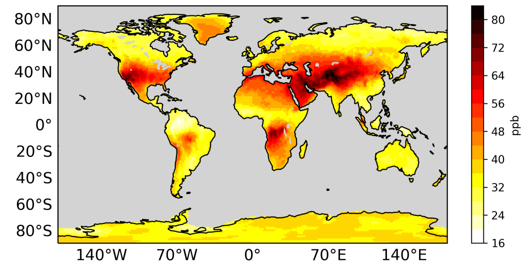
However, the overall decrease in ozone concentration dampens the impact of the change in dry deposition flux. In total, the changes by the revised dry deposition scheme increase the multiyear mean (2010-2015) loss of ozone by dry deposition from 946 $\mathrm{Tg\,yr^{-1}}$ to 1001 $\mathrm{Tg\,yr^{-1}}$ ~~for ozone and from 978 to 1032 for odd oxygen ()[2] which is in the reported range (Hu et al., 2017; Young et al., 2018)~~(Young et al., 2018; Hu et al., 2017). Accordingly, (boreal) summer ground-level ozone over land is reduced by up to 12 ppb (24%) peaking over Scandinavia, Asia, central Africa and East Canada (Fig. 6d). In the Northern Hemisphere, also the zonal mean of the tropospheric ozone mixing ratio show a noticeable reduction far from the ground compared to the default scheme (Fig. 9c). This has the potential to reduce the positive bias of tropospheric ozone on the Northern Hemisphere (20 %) reported by Young et al. (2018). ~~The reduction of ground-level ozone~~ However, besides ozone also other atmospheric tracer gases are affected by the change in dry deposition. The global annual dry deposition flux of odd oxygen (O_x)[2], which includes many important troposheric trace gases, increases from 978 $\mathrm{Tg\,yr^{-1}}$ to 1032 $\mathrm{Tg\,yr^{-1}}$ due to the ~~change in dry deposition is a combined effect of the impact on ozone deposition~~ revision. This is in good agreement with the reported numbers by Hu et al. (2017) and Young et al. (2018). In Fig. 8, we show additionally the absolute and relative change

[2] ~~≡ +++2+3++++++2+~~

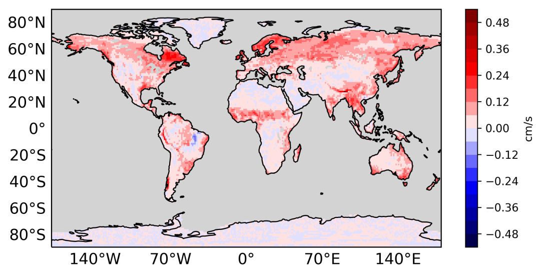
[2] $O_x \equiv O + O_3 + NO_2 + 2NO_3 + 3N_2O_5 + HNO_3 + HNO_4 + BrO + HOBr + BrNO_2 + 2BrNO_3 + PAN$



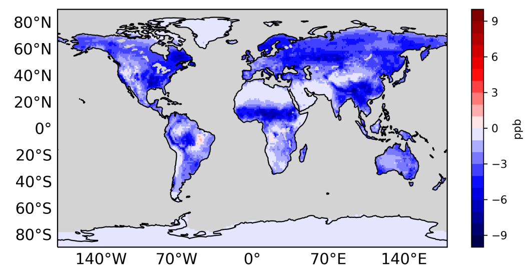
(a) Dry deposition velocity (DEF)



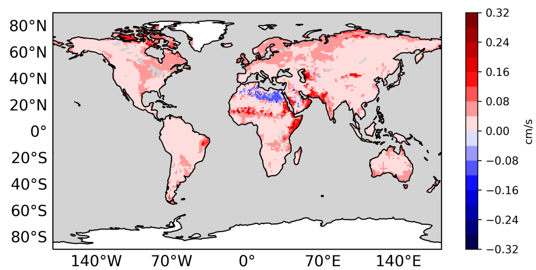
(b) Ground-level ozone (DEF)



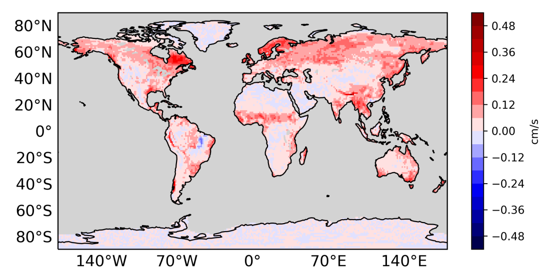
(c) ΔV_d (REV - DEF)



(d) Δ O_3 (REV - DEF)



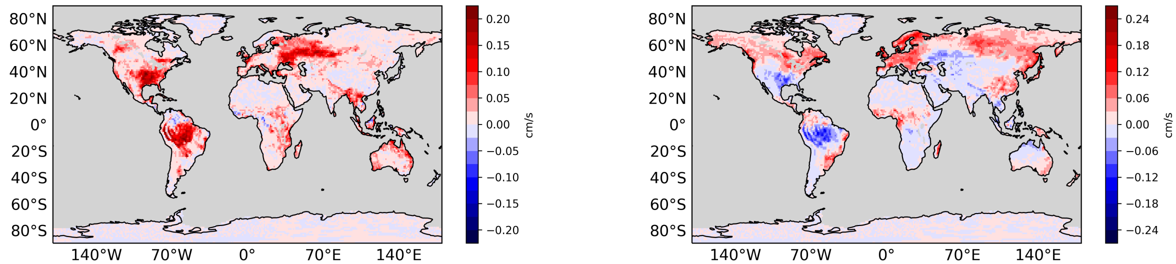
(e) $\Delta V_{cut,d}$ (REV - DEF)



(f) $\Delta V_{cut,w}$ (REV - DEF)

Figure 6. Multiyear (2010-2015) mean absolute values and changes in boreal summer: i.e, difference between revised and default scheme (REV-DEF).

of the multi-year annual average dry deposition loss of SO_2, NO_2,HNO_3 and ~~the~~ HCHO. As a very soluble species the loss of SO_2 is increased by the revised dry deposition scheme whereas the predefined low cuticular and wet skin resistance of HNO_3 in the old scheme were replaced with the new mechanism leading to an decrease in dry deposition. The altered loss of ~~soluble oxygenated VOCs which are~~ NO_2 and HCHO and other ozone precursors at ground level, especially soluble oxygenated VOCs contributes to the total change in ozone loss. NO_2 is deposited almost 40 % more significantly contributing to the net reduction



(a) ΔV_d (REV - REV-fws)

(b) ΔV_d (stress factor inclusion REV - REV-fTfD)

Figure 7. Mean changes (2010) of dry deposition velocity in boreal summer. (a) $f(W_s)$ modification, (b) Temperature and VPD stress

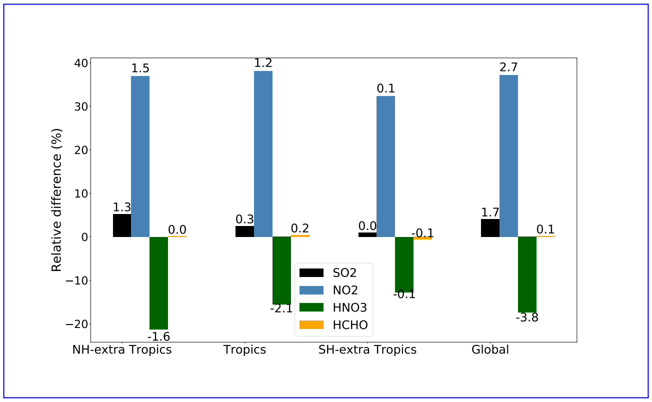
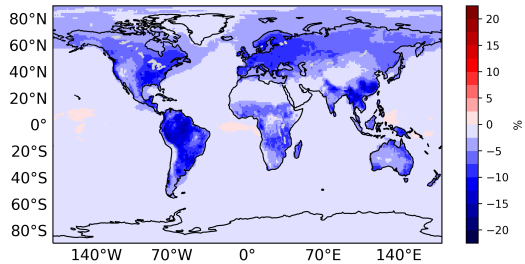


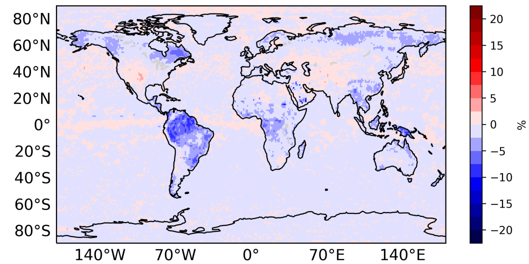
Figure 8. Relative change [%] and absolute change [Tg/yr] (numbers on bars) of annual global loss by dry deposition of O_3, SO_2,HNO_3,HCHO (REV-DEF)

in ozone production but is mostly counterbalanced by other processes. The change of ~~dry deposition~~ HCHO dry deposition flux is small on a global and annual scale and only important regionally, most in (boreal) summer, when it decreases HCHO at ground level (Fig.10b) by up to 25 %~~in (boreal) summer when~~. Thereby, the change in wet uptake is highest but is partially counterbalanced by other effects. This leads to lower HO_2-production from HCHO photooxidation and lower NO-to-NO_2 conversion and thus lower ozone production (Seinfeld and Pandis, 2016). These effects also impact the OH mixing ratio (Fig. 9b, 9d) which control the methane lifetime predicted by the model. However, for a clearer effect, a longer simulated time period would be needed. A detailed analysis of the trace gas budgets is beyond the scope of this manuscript and will be investigated in a subsequent study.

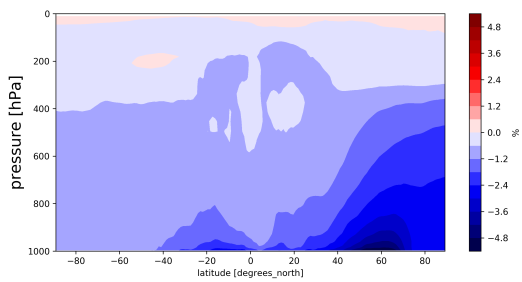
6 Uncertainties in modelling stomatal conductance



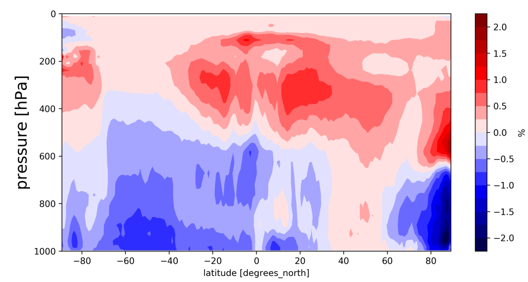
(a) Ground-level ozone mixing ratio



(b) Ground-level OH mixing ratio

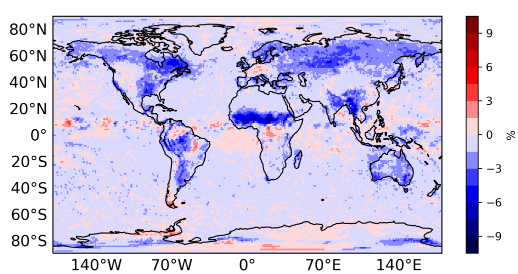


(c) Zonal mean of ozone mixing ratio

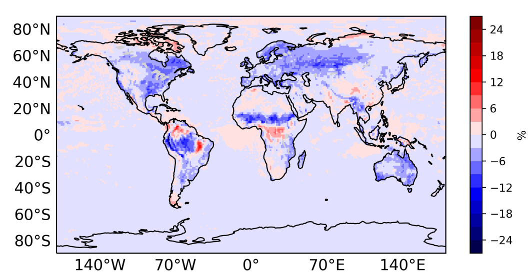


(d) Zonal mean of OH mixing ratio

Figure 9. Relative change of multiyear (2010-2015) mean (DEF-REV)



(a) HO_2 at ground level



(b) HCHO at ground level

Figure 10. Relative change of multiyear (2010-2015) boreal summer mean (DEF-REV)

Dry deposition is a highly uncertain term in modelling ozone pollution (Young et al., 2018; Clifton et al., 2020a). Its representation is general limited by a lack of measurements and process understanding but also too a large extent driven by the quality of land cover information (Hardacre et al., 2015; Clifton et al., 2020b). Although the dry deposition scheme by Wesely (1989) is commonly used in global and regional models (e.g. MOZART, GEOS-Chem) the approach has some constraints (Hardacre et al., 2015)

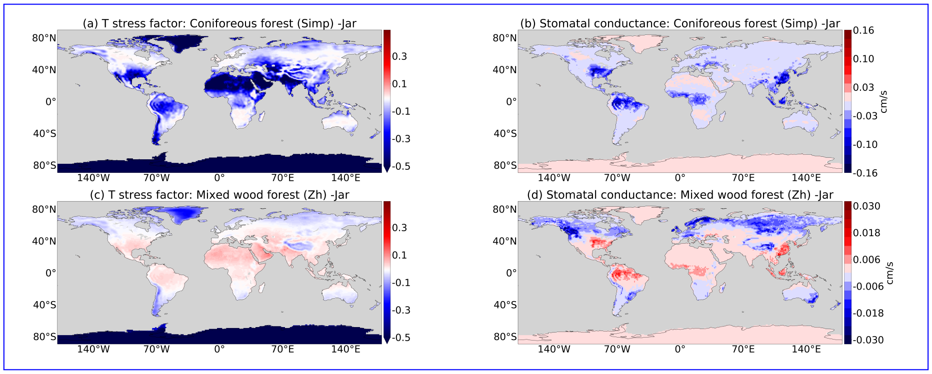
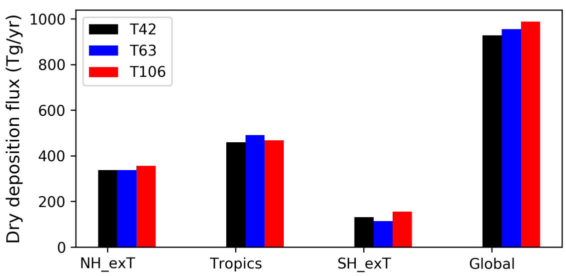


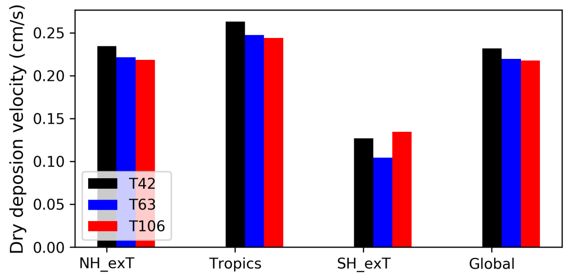
Figure 11. Absolute difference of stomatal conductance applied with the temperature stress factor calculated for two different parameter sets by Simpson et al. (2012) (Simp) and Zhang et al. (2003) (Zh) ein comparison with the here used parameter set by Jarvis (1976) (Jar)

. The disadvantage of the big leaf approach used in MESSy is that a vertical variation of leaf properties, affecting for instance the attenuation of solar radiation is not considered (e.g., Clifton et al., 2020b). Regarding stomatal uptake, we neglect the mesophyll resistance as reactions inside the leaf are commonly assumed to not limit stomatal ozone uptake whereas, besides mostly supporting laboratory studies (e.g., Sun et al., 2016), a few contradicting findings exist (e.g., Tuzet et al., 2011). The here used empirical multiplicative algorithm by Jarvis (1976) for stomatal modelling has one general drawback concerning that the environmental responses to stomata are treated clearly in contrast to experimental evidence (Damour et al., 2010). However, Jarvis-type models have been shown to be able to compete with the semi-mechanistic $A_{net} - g_s$ models which link stomatal uptake to the CO_2 assimilation during plant photosynthesis (Fares et al., 2013; Lu, 2018). The critics in Fares et al. (2013) that the Jarvis model cannot capture the afternoon depression of ozone dry deposition is due to the original used VPD stress factor which has been replaced here by a mechanistic one based on the optimised exchange of CO_2 and water by plants (Katul et al., 2009). Furthermore, a larger set of land cover types is expected to improve the vegetation dependent variation of dry deposition. The parameters used to model dry deposition of stomata, cuticle and soil are biome-dependent and using generalized ones like for the input cuticular resistance can lead to differences in dry deposition (Hoshika et al., 2018) . Exemplary, discrepancies for the stomatal conductance calculated with different parameter sets are shown in Fig. 11 as summer mean of 2010. Thereby, the temperature stress factor have been calculated as in Eq. 6 using the obtained surface temperature by EMAC (Fig. 11 (a),(c)) and applied to the model (DEFAULT) stomatal conductance (Eq. 17) with two different parameter sets for coniferous and mixed forest by Simpson et al. (2012)[3] and Zhang et al. (2003)[4]. Jarvis (1976) obtained the parameters from a set of measurements in mixed hardwood/coniferous forest in Washington. In general, the parameters are related to measurements where the absolute values are influenced by multiple factors like genotype and local climatic conditions (Sulis et al., 2015; Tuovinen et al., 2009; Hoshika et al., 2018). So, for global modelling mostly simplified parameters have to be used like in EMEP (Simpson et al., 2012).

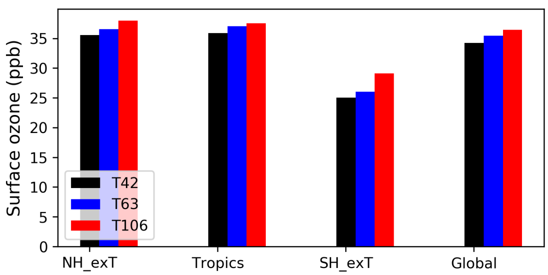
[3]used parameters:$T_{min} = 0°C$,$T_{opt} = 18°C$,$T_{max} = 36°C$
[4]used parameters:$T_{min} = -3°C$,$T_{opt} = 21°C$,$T_{max} = 42°C$



(a) Annual dry deposition loss



(b) Annual mean dry deposition velocity



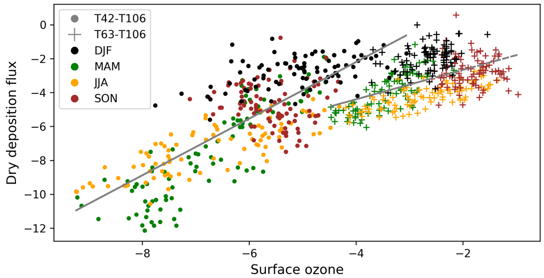
(c) Annual mean surface ozone mixing ratio

Figure 12. Ozone and dry deposition at three different resolutions (T42: 2.8° x2.8°, T63: 1.9° x1.9°, T106: 1.1° x1.1°) and the different regions: Northern Hemisphere extra-Tropics (NH_exT : $90°N - 30°N$), Tropics ($30°N - 30°S$), Southern Hemisphere extra Tropics (SH_exT : $90°S - 30°S$) and the whole Earth (Global).

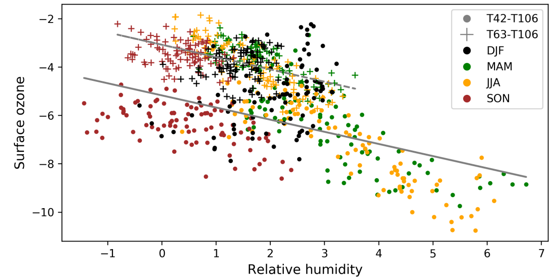
7 Sensitivity to model resolution

The simulation of dry deposition depends on meteorology including boundary layer processes, radiation (cloud distribution and reflectivity) and ozone chemistry as well as on input fields like vegetation density (LAI) (Jones, 1992). Model horizontal resolution inherently affects the amplitude and distribution of (regridded) surface processes and the artificial dilution of ozone precursors that are emitted. This aspect is investigated here by analysing simulations at three different spatial resolutions: 2.8° x2.8°, 1.9° x1.9° and 1.1° x1.1° (REST42, REST63, REV (T106) in Tab. 1).

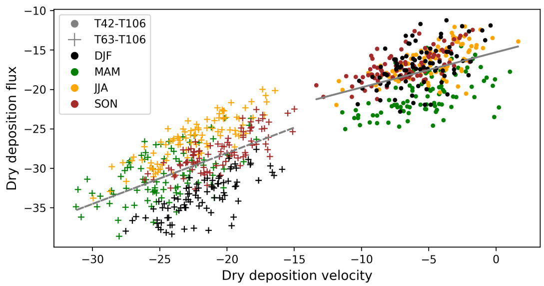
In Figure 12a the resolution dependency is shown for the annual dry deposition flux of ozone on different continental regions. The annual dry deposition fluxes differ by up to 40 $\mathrm{Tg\,yr^{-1}}$ globally between the different resolutions, with highest dry deposition at high resolution (T106). For the Northern Hemisphere (and consequently globally), this difference is driven by the higher annual mean ground-level ozone compared to the lower resolutions (Fig. 12c). However, this effect cannot be disentangled from the effect of decreased dry deposition velocity on ground-level ozone. Globally, increasing differences in O_3 are anti-correlated with relative humidity as shown in Figure 13b ($\rho = -0.8$). The impact of humidity on ozone chemistry is considered to be relatively weak (Jacob and Winner, 2009), but Kavassalis and Murphy (2017) showed for the U.S. that only



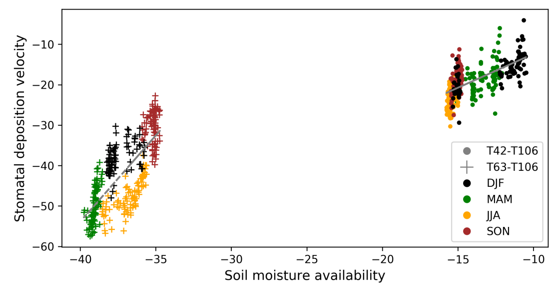
(a) Global: Ground-level ozone and dry deposition flux



(b) Global: Ground-level ozone and relative humidity



(c) SH_exT: Dry deposition flux and velocity



(d) SH_exT: Stomatal dry deposition velocity and soil moisture

Figure 13. Correlations of resolution dependent relative differences of ozone, dry deposition and meteorological variables for the whole Earth (global) ans the Southern Hemisphere extra-Tropics (SH_exT) for the four boreal seasons: spring (MAM), summer (JJA), autumn (SON), winter (DJF).

dry deposition establishes the observed anti-correlation between ozone and relative humidity. A dominating positive correlation of the dry deposition flux with the velocity only occurs on the Southern Hemisphere extra-Tropics (SH_exT), which is highest between T63 and T106 (Fig. 13c). This can be attributed to discrepancies in stomatal deposition (Fig. 13d) driven by differences in humidity which might be caused by different moisture cycles and transpiration.

8 Conclusion and Recommendations

Dry deposition to the Earth's surface is a key process for the representation of ground-level ozone in global models. Its parametrisations constitutes a relevant part of the model uncertainty (Hardacre et al., 2015; Wu et al., 2018). Revising the dry deposition scheme of EMAC leads to an improved representation of surface ozone in regions with a positive model ozone bias (e.g. Europe). The highest increase in ozone dry deposition is due to the implementation of cuticular uptake whose contribution is important especially during night over moist surfaces. The extension of the stomatal uptake with temperature and VPD adjustment factors accounts for the desired link of plant activity to hydroclimate as recommended by Lin et al. (2019).

Especially in drought stressed regions (e.g. Citrus Orchard), the dependence on vapour pressure deficit leads to a realistic depression of stomatal uptake at noon. Also the dependence of dry deposition on soil moisture have been modified since the current representation of soil moisture in the model is not satisfactory. Specifically, the model simulates a too dry soil for the Amazon basin causing stomatal closure and, thus an underestimation of dry deposition (Sect. 4.2). We have indications that the dry bias is a consequence of meteorological nudging in EMAC and also the missing representation of organised convection in the tropics (Mauritsen and Stevens, 2015). The sensitivity of the vegetation to droughts is comparably high in the Amazon region because the model soil cannot hold water in the catchment for a realistic time period and exhibits a memory effect (Hagemann and Stacke, 2013). Deeper root zones or buffering of the soil moisture below the root zone would improve the water holding capacity (Hagemann and Stacke, 2013; Fisher et al., 2007). With an improved representation of soil moisture the more realistic parametrisation of the soil moisture stress on stomatal uptake could be re-enabled. In general, the inclusion of the strong link between dry deposition and meteorology reveals some limitations of the dry deposition scheme associated with the inaccurate representation of local meteorology. The results also indicate that an improved representation of important non-stomatal dry deposition like in-canopy reactions of ozone with volatile organic compounds (e.g. Citrus Orchard, Sect. 4.2) would lower the positive model-observation discrepancy. This can be achieved with the inclusion of further biogenic VOCs and an explicit parametrization of the transport dynamics in the boundary layer in model simulations (Makar et al., 2017). Explicit field measurements could foster further process understanding, which is required for a detailed process description within the models, especially over tropical rain-forests. The seasonal variability of the simulated dry deposition velocity could be further improved by using as model input the time-series of vegetation cover from an imaging products which also capture land use changes and vegetation trend that are known to impact dry deposition significantly (Wong et al., 2019).

9 Outlook

The representation of gaseous dry deposition in MESSy will be further improved by using the MODIS time-series of LAI which captures multi-annual vegetation changes. As the next step of dry deposition modelling in MESSy a biome-dependent dry deposition model coupled to CO_2 assimilation (White et al. 2004) will be applied. Biome-dependent vegetation cover information, required for this scheme, are then provided by global input data which, however, represent only the annual cycle of vegetation. Coupling MESSy to the recently available dynamic vegetation model LPJ-GUESS providing detailed vegetation information with the temporal variability required for a climate model could be a further improvement. By now the one-way coupling of LPJ-GUESS as a MESSy submodel is only in the initial evaluation phase of the coupling with the atmospheric model (Forrest et al., 2020).

Data availability. The measurement data at Ontario is freely available at http://data.ec.gc.ca/data/air/monitor/special-studies-of-atmospheric-gases-particles-and-precipitation-chemistry/borden-forest-ozone-and-sulphur-dioxide-dry-deposition-study with the 'Open Gouvernment Licence-Canada' (https://open.canada.ca/en/open-government-licence-canada). The measurement data at Hytiälä (Creative Commons 4.0 Attribution

(CC BY) license https://creativecommons.org/licenses/by/4.0/) can be accessed at https://avaa.tdata.fi/web/smart/smear/download. The data from Lindcove station (Fares) were provided by S. Fares (Fares et al., 2012). The dry deposition measurement data at Amazonian Tall Tower Observatory was provided by Matthias Sörgel and is available on request. The used global climate reanalysis ERA5 by ECMWF are available through the Climate Data Store (https://cds.climate.copernicus.eu).

Code availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licenced to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Website http://www.messy-interface.org. The code presented here has been based on MESSy version 2.54 and will be available in the next official release (version 2.55). The exact code version used to produce the results of this paper is archived in the MESSy code repository and can be made available to members of the MESSy community upon request.

Appendix A: Default dry deposition scheme

The default dry deposition scheme of MESSy uses the following equations described in Kerkweg et al. (2006).

Surface resistance over vegetation (in $\mathrm{s\,m^{-1}}$):

$$\frac{1}{R_{s,veg}(X)} = \frac{1}{R_{can} + R_{s,soil}(X) + R_{qbr,veg}(X)} + \frac{LAI}{r_{cut}(X)} + \frac{LAI}{r_{stom,corr}(X) + r_{mes}(X)} \tag{A1}$$

where $R_{can}(X)$, $R_{s,soil}(X)$, $R_{qbr,veg}(X)$ are the in-canopy aerodynamic resistance, the soil resistance and the quasi-laminar boundary resistance at canopy scale (in $\mathrm{s\,m^{-1}}$). $r_{cut}(X)$, $r_{stom,corr}(X)$ and $r_{mes}(X)$ are the cuticular resistance, stomatal resistance and mesophyll resistance at leaf scale scaled with Leaf Area Index (LAI in $\mathrm{m^2\,m^{-2}}$) to canopy scale.

Stomatal resistance:

$$r_{stom,corr} = \frac{r_{stom}(PAR)}{fws} \cdot \frac{D_{\mathrm{H_2O}}}{D(\mathrm{O_3})} \tag{A2}$$

Soil moisture stress function:

$$f(W_s) = \begin{cases} 1 & W_s(t) \geq W_{cr} (= 75\%) \\ \frac{W_s(t) - W_{pwp}}{W_{cr} - W_{pwp}} & W_{pwp} < W_s(t) < W_{cr} \\ 0 & W_s(t) \leq W_{pwp} (= 35\%) \end{cases} \tag{A3}$$

Cuticular resistance:

$$r_{cut}(X) = \frac{r_{cut}(\mathrm{O_3})}{10^{-5} \cdot H(\mathrm{O_3}) + s_{reac}(\mathrm{O_3})} \tag{A4}$$

where $r_{cut}\mathrm{O_3}$=$1\mathrm{e}-5\ \mathrm{s\,m^{-1}}$, H($\mathrm{O_3}$)=0.01 and s_{reac}=1.

Wet skin resistance:

$$R_{ws}(\mathrm{O_3}) = \left[\frac{1/3}{R_{ws}(\mathrm{SO_2})} + 10^{-7} \cdot H(\mathrm{O_3}) + \frac{s_{reac}(\mathrm{O_3})}{R_{cut,w}(\mathrm{O_3})}\right]^{-1} \tag{A5}$$

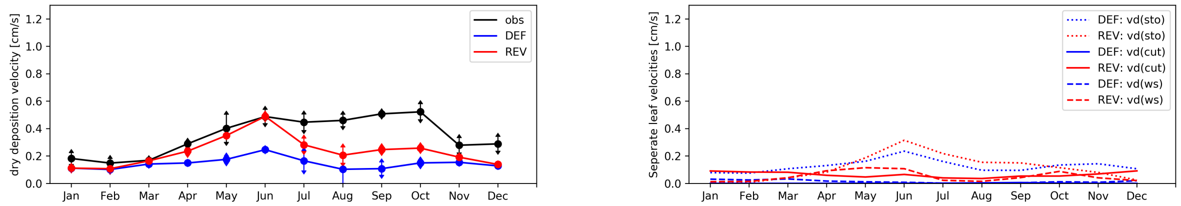
where R_{ws}($\mathrm{O_3}$)=2000 $\mathrm{s\,m^{-1}}$ and R_{ws}($\mathrm{SO_2}$)=100 $\mathrm{s\,m^{-1}}$.

Appendix B: Evapotranspiration

Plants play a key role in the water and energy cycle and thus contribute to the land-atmosphere coupling, which drives the global climate. In this context, transpiration is an important process, as plants loose water during the necessary CO_2 uptake via their stomata. The amount depends on the aperture behaviour of the respective plant in the respective environmental conditions (Katul et al., 2012). Thus, the latent heat flux incorporates the canopy resistance. The formulation is based on the Monin-Obukov stability theory:

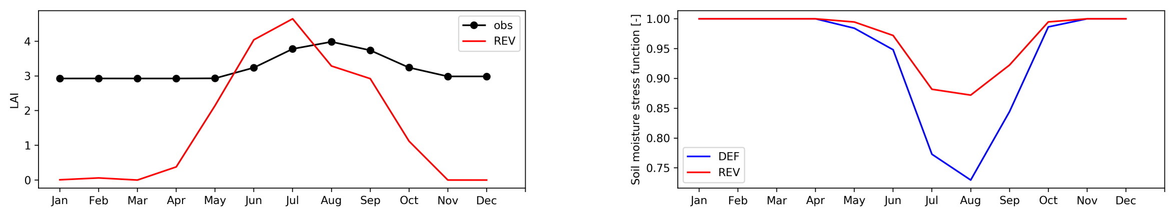
$$E = \rho C_h |\boldsymbol{v}| \beta (q_a - h q_s(T_s, p_s)) \qquad \beta = \left[1 + \frac{C_h |\boldsymbol{v}| R_{stom}}{fws}\right]^{-1} \tag{B1}$$

where ρ is the density of air, $|\boldsymbol{v}|$ is the absolute value of the horizontal wind speed, C_h is the transfer coefficent of heat whereas $r_a = 1/(C_h|\boldsymbol{v}|)$. q_s and q_a are the saturation-specific humidity and the atmospheric specific humidity whereas the relative humidity h at the surface limits the evapotranspiration from bare soil. β determines the ratio of transpiration between water stressed plants (β <1) and well-watered plants (β =1) (Giorgetta et al., 2013; Schulz et al., 2001). The formular for the canopy stomatal resistance R_{stom} is given in Eq. 5. In order to adapt the transpiration to temperature and vapour pressure deficit the T and VPD adjustment factors can be applied to R_{stom} inversely like in the new dry deposition scheme via $izwet = 1$ in the VERTEX $\&CTRL$ namelist. The modification of the soil moisture stress function $f(W_s)$ (old: Eq. A, new: Eq. 12) affects evapotranspiration directly.

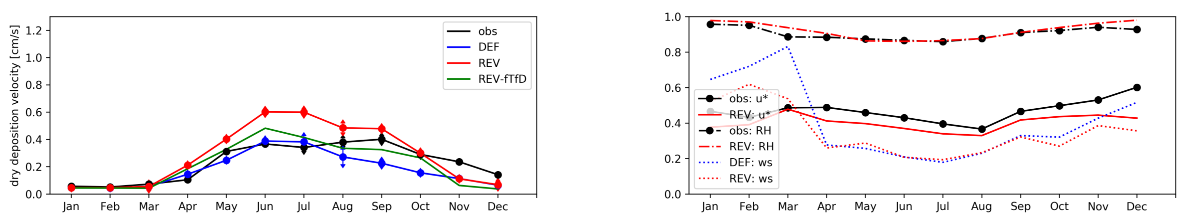


(a) Total dry deposition velocity (night-time mean) (b) Contributing leaf deposition velocities (night-time mean)

Figure A1. Measured and modelled (DEF, REV) annual cycle at Borden forest



(a) Leaf area index (b) Soil moisture stress function



(c) Dry deposition velocity at RH>70% (d) Friction velocity, relative humidity, wet skin fraction at RH>70%

Figure A2. Measured (obs) and modelled (DEF, REV) multiyear (2010-2012) and REV-fTfD (2010) annual cycle at Hyytiälä

Author contributions. D.T. (and A.K.) initiated and supervised the study. D.T. and T.E. discussed the model developments which were implemented by A.K. and T.E.. H.O. originally wrote the MESSy vertical diffusion submodel VERTEX. S.F. provided the measurement data from Lindcove and further related theoretical calculations. I.M. did the dry deposition measurements at Hytiälä and gave related support. T.E. performed the EMAC simulations, the data analyses, prepared the figures and wrote the manuscript.

Competing interests. The authors declare no competing financial interests.

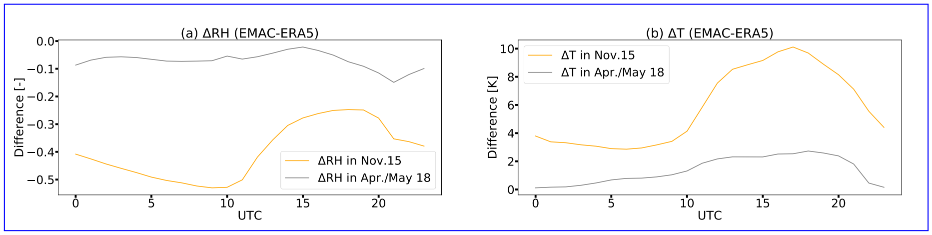
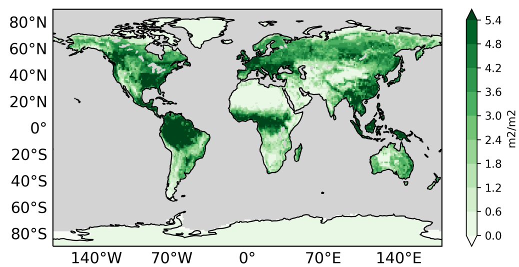
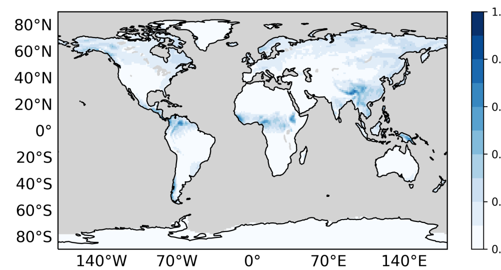


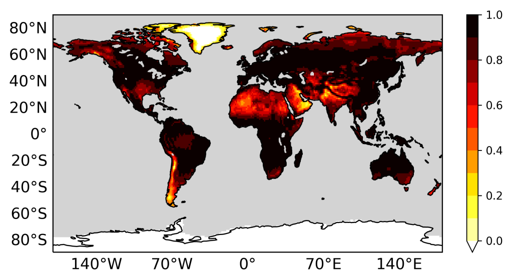
Figure A3. Differences of meteorology between EMAC and ERA5 at ATTO



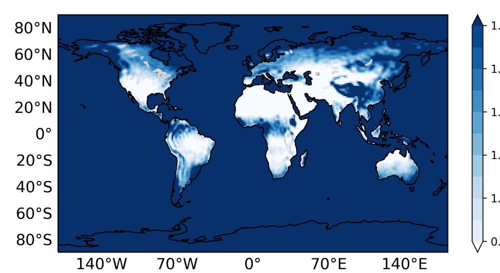
(a) Leaf Area Index



(b) Wet skin fraction



(c) Temperature stress factor



(d) VPD stress factor

Figure A4. Boreal summer mean vegetation and meteorological variables predicted by EMAC

Acknowledgements. The work described in this paper has received funding from the Initiative and Networking Fund of the Helmholtz Association through the project "Advanced Earth System Modelling Capacity (ESM)". The content of this paper is the sole responsibility of the author(s) and it does not represent the opinion of the Helmholtz Association, and the Helmholtz Association is not responsible for any use that might be made of the information contained. The author(s) acknowledge the Environment and Climate Change Canada and the United States Environmental Protection Agency for the provision of the dry deposition velocity data at the Borden forest measurement station. Moreover, the personnel at SMEAR II station of INAR – Institute for Atmospheric and Earth System Research, University of Helsinki, Finland, is acknowledged. Concerning the measurement data from Amazonian Tall Tower, we thank the Instituto Nacional de Pesquisas da Amazonia (INPA) and the Max Planck Society for continuous support. We thank for the support by the German Federal Ministry of Education and Research (BMBF contracts 01LB1001A, 01LK1602B and 01LP1606B) and the Brazilian Ministério da Ciência, Tecnologia e Inovação (MCTI/FINEP contract 01.11.01248.00) as well as the Amazon State University (UEA), FAPEAM, LBA/INPA and SDS/CEUC/RDS-Uatumã. The measurements were conducted by Matthias Sörgel, Anywhere Tsokankunku, Stefan Wolff and Rodrigo Souza. For the usage of data from the ERA5 global climate reanalysis (Generated using Copernicus Atmosphere Monitoring Service Information [2020]) we acknowledge the Copernicus Climate Change and Atmosphere Monitoring Service (https://apps.ecmwf.int/datasets/licences/copernicus/). Neither the European Commission nor ECMWF is responsible for any use that may be made of the Copernicus information or data it contains.

References

Altimir, N., Kolari, P., Tuovinen, J.-P., Vesala, T., Bäck, J., Suni, T., Kulmala, M., and Hari, P.: Foliage surface ozone deposition: a role for surface moisture?, 2006.

Andersson, C. and Engardt, M.: European ozone in a future climate: Importance of changes in dry deposition and isoprene emissions, Journal of Geophysical Research: Atmospheres, 115, https://doi.org/10.1029/2008JD011690, 2010.

Baldocchi, D. D., Hicks, B. B., and Camara, P.: A canopy stomatal resistance model for gaseous deposition to vegetated surfaces, Atmospheric Environment (1967), 21, 91–101, 1987.

Ball, J. T., Woodrow, I. E., and Berry, J. A.: A model predicting stomatal conductance and its contribution to the control of photosynthesis under different environmental conditions, in: Progress in photosynthesis research, pp. 221–224, Springer, 1987.

Bourtsoukidis, E., Behrendt, T., Yañez-Serrano, A. M., Hellén, H., Diamantopoulos, E., Catão, E., Ashworth, K., Pozzer, A., Quesada, C., Martins, D., et al.: Strong sesquiterpene emissions from Amazonian soils, Nature communications, 9, 1–11, https://doi.org/10.1038/s41467-018-04658-y, 2018.

Clifton, O., Paulot, F., Fiore, A., Horowitz, L., Correa, G., Baublitz, C., Fares, S., Goded, I., Goldstein, A., Gruening, C., et al.: Influence of dynamic ozone dry deposition on ozone pollution, Journal of Geophysical Research: Atmospheres, 125, e2020JD032 398, https://doi.org/10.1029/2020JD032398, 2020a.

Clifton, O. E., Fiore, A. M., Munger, J., Malyshev, S., Horowitz, L., Shevliakova, E., Paulot, F., Murray, L., and Griffin, K.: Interannual variability in ozone removal by a temperate deciduous forest, Geophysical Research Letters, 44, 542–552, https://doi.org/10.1002/2016GL070923, 2017.

Clifton, O. E., Fiore, A. M., Massman, W. J., Baublitz, C. B., Coyle, M., Emberson, L., Fares, S., Farmer, D. K., Gentine, P., Gerosa, G., et al.: Dry deposition of ozone over land: processes, measurement, and modeling, Reviews of Geophysics, 58, e2019RG000 670, https://doi.org/10.1029/2019RG000670, 2020b.

Collatz, G. J., Ribas-Carbo, M., and Berry, J.: Coupled photosynthesis-stomatal conductance model for leaves of C4 plants, Functional Plant Biology, 19, 519–538, 1992.

Combe, M., de Arellano, J. V.-G., Ouwersloot, H. G., and Peters, W.: Plant water-stress parameterization determines the strength of land–atmosphere coupling, Agricultural and forest meteorology, 217, 61–73, https://doi.org/10.1016/j.agrformet.2015.11.006, 2016.

Copernicus, C. C. C. S. C.: ERA5: Fifth generation of ECMWF atmospheric reanalyses of the global climate, https://cds.climate.copernicus.eu/cdsapp#!/home, https://doi.org/10.24381/cds.adbb2d47, accessed April 8th, 2020, 2017.

Coumou, D. and Rahmstorf, S.: A decade of weather extremes, Nature climate change, 2, 491, https://doi.org/0.1038/NCLIMATE1452, 2012.

Cowan, I. and Farquhar, G.: Stomatal function in relation to leaf metabolism and environment., 1977.

Damour, G., Simonneau, T., Cochard, H., and Urban, L.: An overview of models of stomatal conductance at the leaf level, Plant, Cell & Environment, 33, 1419–1438, https://doi.org/0.1111/j.1365-3040.2010.02181.x, 2010.

Deckert, R., Jöckel, P., Grewe, V., Gottschaldt, K.-D., and Hoor, P.: A quasi chemistry-transport model mode for EMAC, Geoscientific Model Development, 4, 195, https://doi.org/10.5194/gmd-4-195-2011, 2011.

Delworth, T. L. and Manabe, S.: The influence of potential evaporation on the variabilities of simulated soil wetness and climate, Journal of Climate, 1, 523–547, 1988.

Emberson, L., Ashmore, M., Cambridge, H., Simpson, D., and Tuovinen, J.-P.: Modelling stomatal ozone flux across Europe, Environmental Pollution, 109, 403–413, 2000.

Fares, S.: Measurement data at Lindcove Orange Orchard, https://doi.org/10.18140/FLX/1440233.

Fares, S., Weber, R., Park, J.-H., Gentner, D., Karlik, J., and Goldstein, A. H.: Ozone deposition to an orange orchard: Partitioning between stomatal and non-stomatal sinks, Environmental Pollution, 169, 258–266, https://doi.org/10.1016/j.envpol.2012.01.030, 2012.

Fares, S., Matteucci, G., Mugnozza, G. S., Morani, A., Calfapietra, C., Salvatori, E., Fusaro, L., Manes, F., and Loreto, F.: Testing of models of stomatal ozone fluxes with field measurements in a mixed Mediterranean forest, Atmospheric environment, 67, 242–251, https://doi.org/10.1016/j.atmosenv.2012.11.007, 2013.

Fisher, R., Williams, M., Da Costa, A. L., Malhi, Y., Da Costa, R., Almeida, S., and Meir, P.: The response of an Eastern Amazonian rain forest to drought stress: results and modelling analyses from a throughfall exclusion experiment, Global Change Biology, 13, 2361–2378, https://doi.org/10.1111/j.1365-2486.2007.01417.x, 2007.

Freire, L., Gerken, T., Ruiz-Plancarte, J., Wei, D., Fuentes, J., Katul, G., Dias, N., Acevedo, O., and Chamecki, M.: Turbulent mixing and removal of ozone within an Amazon rainforest canopy, Journal of Geophysical Research: Atmospheres, 122, 2791–2811, https://doi.org/10.1002/2016JD026009, 2017.

Ganzeveld, L. and Lelieveld, J.: Dry deposition parameterization in a chemistry general circulation model and its influence on the distribution of reactive trace gases, Journal of Geophysical Research: Atmospheres, 100, 20 999–21 012, 1995.

Ganzeveld, L., Lelieveld, J., and Roelofs, G.-J.: A dry deposition parameterization for sulfur oxides in a chemistry and general circulation model, Journal of Geophysical Research: Atmospheres, 103, 5679–5694, 1998.

Ganzeveld, L., Lelieveld, J., Dentener, F., Krol, M., Bouwman, A., and Roelofs, G.-J.: Global soil-biogenic NOx emissions and the role of canopy processes, Journal of Geophysical Research: Atmospheres, 107, 2002.

Giorgetta, M. A., Roeckner, E., Mauritsen, T., Bader, J., Crueger, T., Esch, M., Rast, S., Kornblueh, L., Schmidt, H., Kinne, S., et al.: The atmospheric general circulation model ECHAM6-model description, 2013.

Hagemann, S. and Stacke, T.: Impact of the soil hydrology scheme on simulated soil moisture memory in a GCM, in: EGU General Assembly Conference Abstracts, vol. 15, https://doi.org/10.1007/s00382-014-2221-6, 2013.

Hardacre, C., Wild, O., and Emberson, L.: An evaluation of ozone dry deposition in global scale chemistry climate models, Atmospheric Chemistry and Physics, 15, 6419–6436, https://doi.org/10.5194/acp-15-6419-2015, 2015.

Hogg, A., Uddling, J., Ellsworth, D., Carroll, M. A., Pressley, S., Lamb, B., and Vogel, C.: Stomatal and non-stomatal fluxes of ozone to a northern mixed hardwood forest, Tellus B: Chemical and Physical Meteorology, 59, 514–525, https://doi.org/10.1111/j.1600-0889.2007.00269.x, 2007.

Hoshika, Y., Osada, Y., De Marco, A., Penuelas, J., and Paoletti, E.: Global diurnal and nocturnal parameters of stomatal conductance in woody plants and major crops, Global ecology and biogeography, 27, 257–275, https://doi.org/10.1016/j.agrformet.2017.01.005, 2018.

Hu, L., Jacob, D. J., Liu, X., Zhang, Y., Zhang, L., Kim, P. S., Sulprizio, M. P., and Yantosca, R. M.: Global budget of tropospheric ozone: Evaluating recent model advances with satellite (OMI), aircraft (IAGOS), and ozonesonde observations, Atmospheric Environment, 167, 323–334, https://doi.org/10.1016/j.atmosenv.2017.08.036, 2017.

Huang, L., McDonald-Buller, E. C., McGaughey, G., Kimura, Y., and Allen, D. T.: The impact of drought on ozone dry deposition over eastern Texas, Atmospheric environment, 127, 176–186, https://doi.org/10.1016/j.atmosenv.2015.12.022, 2016.

Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, Atmospheric environment, 43, 51–63, https://doi.org/10.1016/j.atmosenv.2008.09.051, 2009.

Jarvis, P.: The interpretation of the variations in leaf water potential and stomatal conductance found in canopies in the field, Phil. Trans. R. Soc. Lond. B, 273, 593–610, 1976.

Jeuken, A. B. M., Siegmund, P. C., Heijboer, L. C., Feichter, J., and Bengtsson, L.: On the potential of assimilating meteorological analyses in a global climate model for the purpose of model validation, Journal of Geophysical Research: Atmospheres, 101, 16 939–16 950, https://doi.org/10.1029/96JD01218, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/96JD01218, 1996.

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the modular earth submodel system (MESSy2), Geoscientific Model Development, 3, 717–752, https://doi.org/10.5194/gmd-3-717-2010, 2010.

Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., et al.: Earth system chemistry integrated modelling (ESCiMo) with the modular earth submodel system (MESSy) version 2.51, https://doi.org/10.5194/gmd-9-1153-2016, 2016.

Jones, H.: Plants and Microclimate, 428 pp, Cambridge University Press, Cambridge, 1992.

Jülich Supercomputing Centre: JURECA: Modular supercomputer at Jülich Supercomputing Centre, Journal of large-scale research facilities, 4, https://doi.org/10.17815/jlsrf-4-121-1, http://dx.doi.org/10.17815/jlsrf-4-121-1, 2018.

Katul, G. G., Palmroth, S., and Oren, R.: Leaf stomatal responses to vapour pressure deficit under current and CO2-enriched atmosphere explained by the economics of gas exchange, Plant, Cell & Environment, 32, 968–979, https://doi.org/10.1111/j.1365-3040.2009.01977.x, 2009.

Katul, G. G., Oren, R., Manzoni, S., Higgins, C., and Parlange, M. B.: Evapotranspiration: a process driving mass transport and energy exchange in the soil-plant-atmosphere-climate system, Reviews of Geophysics, 50, https://doi.org/10.1029/2011RG000366, 2012.

Kavassalis, S. C. and Murphy, J. G.: Understanding ozone-meteorology correlations: A role for dry deposition, Geophysical Research Letters, 44, 2922–2931, https://doi.org/10.1002/2016GL071791, 2017.

Kerkweg, A., Buchholz, J., Ganzeveld, L., Pozzer, A., Tost, H., and Jöckel, P.: An implementation of the dry removal processes DRY DEPosition and SEDImentation in the Modular Earth Submodel System (MESSy), Atmospheric Chemistry and Physics, 6, 4617–4632, 2006.

Keronen, P., Reissell, A., Rannik, U., Pohja, T., Siivola, E., Hiltunen, V., Hari, P., Kulmala, M., and Vesala, T.: Ozone flux measurements over a Scots pine forest using eddy covariance method: performance evaluation and comparison with flux-profile method, Boreal environment research, 8, 425–444, 2003.

Kharol, S., Shephard, M., McLinden, C., Zhang, L., Sioris, C., O'Brien, J., Vet, R., Cady-Pereira, K., Hare, E., Siemons, J., et al.: Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America, Geophysical Research Letters, 45, 1157–1166, https://doi.org/10.1002/2017GL075832, 2018.

Klimarechenzentrum, D. et al.: The ECHAM3 atmospheric general circulation model, Techn. Rep, 6, 1992.

Kraus, H.: Die Atmosphäre der Erde: Eine Einführung in die Meteorologie, Springer-Verlag, 2007.

Lamaud, E., Carrara, A., Brunet, Y., Lopez, A., and Druilhet, A.: Ozone fluxes above and within a pine forest canopy in dry and wet conditions, Atmospheric Environment, 36, 77–88, 2002.

Lamaud, E., Loubet, B., Irvine, M., Stella, P., Personne, E., and Cellier, P.: Partitioning of ozone deposition over a developed maize crop between stomatal and non-stomatal uptakes, using eddy-covariance flux measurements and modelling, Agricultural and Forest Meteorology, 149, 1385–1396, https://doi.org/10.1016/j.agrformet.2009.03.017, 2009.

Lin, M., Malyshev, S., Shevliakova, E., Paulot, F., Horowitz, L. W., Fares, S., Mikkelsen, T. N., and Zhang, L.: Sensitivity of ozone dry deposition to ecosystem-atmosphere interactions: A critical appraisal of observations and simulations, Global Biogeochemical Cycles, https://doi.org/10.1029/2018GB006157, 2019.

Lu, Y.-S.: Propagation of land surface model uncertainties in simulated terrestrial system states, 2018.

Makar, P., Staebler, R., Akingunola, A., Zhang, J., McLinden, C., Kharol, S., Pabla, B., Cheung, P., and Zheng, Q.: The effects of forest canopy shading and turbulence on boundary layer ozone, Nature communications, 8, 15 243, 2017.

Mauritsen, T. and Stevens, B.: Missing iris effect as a possible cause of muted hydrological change and high climate sensitivity in models, Nature Geoscience, 8, 346–351, https://doi.org/0.1038/NGEO2414, 2015.

Mauritsen, T., Stevens, B., Roeckner, E., Crueger, T., Esch, M., Giorgetta, M., Haak, H., Jungclaus, J., Klocke, D., Matei, D., et al.: Tuning the climate of a global model, Journal of advances in modeling Earth systems, 4, https://doi.org/10.1029/2012MS000154, 2012.

Mészáros, R., Horváth, L., Weidinger, T., Neftel, A., Nemitz, E., Dämmgen, U., Cellier, P., and Loubet, B.: Measurement and modelling ozone fluxes over a cut and fertilized grassland., Biogeosciences, 6, 2009.

National Centers for Environmental Information, N.: State of the Climate: Global Climate Report for Annual 2015, Tech. rep., https://www.ncdc.noaa.gov/sotc/global/201513.

Ran, L., Pleim, J., Song, C., Band, L., Walker, J. T., and Binkowski, F. S.: A photosynthesis-based two-leaf canopy stomatal conductance model for meteorology and air quality modeling with WRF/CMAQ PX LSM, Journal of Geophysical Research: Atmospheres, 122, 1930–1952, https://doi.org/10.1002/2016JD025583, 2017.

Rannik, Ü., Altimir, N., Mammarella, I., Bäck, J., Rinne, J., Ruuskanen, T., Hari, P., Vesala, T., and Kulmala, M.: Ozone deposition into a boreal forest over a decade of observations: evaluating deposition partitioning and driving variables, Atmospheric Chemistry and Physics, 12, 12 165–12 182, https://doi.org/10.5194/acp-12-12165-2012, 2012.

Rannik, Ü., Peltola, O., and Mammarella, I.: Random uncertainties of flux measurements by the eddy covariance technique, Atmospheric Measurement Techniques, 9, 5163, https://doi.org/0.5194/amt-9-5163-2016, 2016.

Righi, M., Eyring, V., Gottschaldt, K.-D., Klinger, C., Frank, F., Jöckel, P., and Cionni, I.: Quantitative evaluation of ozone and selected climate parameters in a set of EMAC simulations, Geoscientific Model Development, 8, 733–768, https://doi.org/10.5194/gmd-8-733-2015, https://www.geosci-model-dev.net/8/733/2015/, 2015.

Roeckner, E., Bäuml, G., Bonaventura, L., Brokopf, R., Esch, M., Giorgetta, M., Hagemann, S., Kirchner, I., Kornblueh, L., Manzini, E., et al.: The atmospheric general circulation model ECHAM 5. PART I: Model description, 2003.

Rummel, U., Ammann, C., Kirkman, G., Moura, M., Foken, T., Andreae, M., and Meixner, F.: Seasonal variation of ozone deposition to a tropical rain forest in southwest Amazonia, 2007.

Schulz, J.-P., Dümenil, L., and Polcher, J.: On the land surface–atmosphere coupling and its impact in a single-column atmospheric model, Journal of Applied Meteorology, 40, 642–663, 2001.

Schwede, D., Zhang, L., Vet, R., and Lear, G.: An intercomparison of the deposition models used in the CASTNET and CAPMoN networks, Atmospheric environment, 45, 1337–1346, 2011.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric chemistry and physics: from air pollution to climate change, John Wiley & Sons, 2016.

Sellers, P., Mintz, Y., Sud, Y. e. a., and Dalcher, A.: A simple biosphere model (SiB) for use within general circulation models, Journal of the Atmospheric Sciences, 43, 505–531, 1986.

Sellers, P., Dickinson, R. E., Randall, D., Betts, A., Hall, F., Berry, J., Collatz, G., Denning, A., Mooney, H., Nobre, C., et al.: Modeling the exchanges of energy, water, and carbon between continents and the atmosphere, Science, 275, 502–509, 1997.

Sellers, P. J.: Canopy reflectance, photosynthesis and transpiration, International Journal of Remote Sensing, 6, 1335–1372, 1985.

Seneviratne, S. I., Corti, T., Davin, E. L., Hirschi, M., Jaeger, E. B., Lehner, I., Orlowsky, B., and Teuling, A. J.: Investigating soil moisture–climate interactions in a changing climate: A review, Earth-Science Reviews, 99, 125–161, https://doi.org/10.1016/j.earscirev.2010.02.004, 2010.

Silva, S. J. and Heald, C. L.: Investigating dry deposition of ozone to vegetation, Journal of Geophysical Research: Atmospheres, 123, 559–573, 2018.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., et al.: The EMEP MSC-W chemical transport model-technical description, https://doi.org/10.5194/acp-12-7825-2012, 2012.

Solberg, S., Hov, Ø., Søvde, A., Isaksen, I., Coddeville, P., De Backer, H., Forster, C., Orsolini, Y., and Uhse, K.: European surface ozone in the extreme summer 2003, Journal of Geophysical Research: Atmospheres, 113, https://doi.org/10.1029/2007JD009098, 2008.

Stella, P., Loubet, B., Lamaud, E., Laville, P., and Cellier, P.: Ozone deposition onto bare soil: a new parameterisation, Agricultural and Forest Meteorology, 151, 669–681, 2011.

Stella, P., Loubet, B., de Berranger, C., Charrier, X., Ceschia, E., Gerosa, G., Finco, A., Lamaud, E., Serça, D., George, C., et al.: Soil ozone deposition: Dependence of soil resistance to soil texture, Atmospheric Environment, 199, 202–209, 2019.

Stephens, G. L., Li, J., Wild, M., Clayson, C. A., Loeb, N., Kato, S., L'ecuyer, T., Stackhouse, P. W., Lebsock, M., and Andrews, T.: An update on Earth's energy balance in light of the latest global observations, Nature Geoscience, 5, 691–696, https://doi.org/10.1038/NGEO1580, 2012.

Stevens, B. and Schwartz, S. E.: Observing and modeling Earth's energy flows, Surveys in geophysics, 33, 779–816, https://doi.org/10.1007/s10712-012-9184-0, 2012.

Sulis, M., Langensiepen, M., Shrestha, P., Schickling, A., Simmer, C., and Kollet, S. J.: Evaluating the influence of plant-specific physiological parameterizations on the partitioning of land surface energy fluxes, Journal of hydrometeorology, 16, 517–533, https://doi.org/10.1175/JHM-D-14-0153.1, 2015.

Sun, S., Moravek, A., von der Heyden, L., Held, A., Sörgel, M., and Kesselmeier, J.: Twin-cuvette measurement technique for investigation of dry deposition of O3 and PAN to plant leaves under controlled humidity conditions, Atmos Meas Tech, 9, 599–617, https://doi.org/10.5194/amt-9-599-2016, 2016.

Tuovinen, J.-P., Emberson, L., and Simpson, D.: Modelling ozone fluxes to forests for risk assessment: status and prospects, Annals of Forest Science, 66, 1–14, https://doi.org/10.1051/forest/2009024, 2009.

Tuzet, A., Perrier, A., Loubet, B., and Cellier, P.: Modelling ozone deposition fluxes: The relative roles of deposition and detoxification processes, Agricultural and Forest Meteorology, 151, 480–492, https://doi.org/10.1016/j.agrformet.2010.12.004, 2011.

Val Martin, M., Heald, C., and Arnold, S.: Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O3, Geophysical Research Letters, 41, 2988–2996, https://doi.org/10.1002/2014GL059651, 2014.

Van Pul, W. and Jacobs, A.: The conductance of a maize crop and the underlying soil to ozone under various environmental conditions, Boundary-Layer Meteorology, 69, 83–99, 1994.

Wang, Y., Sperry, J. S., Anderegg, W. R., Venturas, M. D., and Trugman, A. T.: A theoretical and empirical assessment of stomatal optimization modeling, New Phytologist, https://doi.org/10.1111/nph.16572, 2020.

Wesely, M.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmospheric Environment (1967), 23, 1293–1304, 1989.

Wesely, M. and Hicks, B.: Some factors that affect the deposition rates of sulfur dioxide and similar gases on vegetation, Journal of the Air Pollution Control Association, 27, 1110–1116, 1977.

Wesely, M. and Hicks, B.: A review of the current status of knowledge on dry deposition, Atmospheric environment, 34, 2261–2282, 2000.

Wong, A. Y., Geddes, J. A., Tai, A. P., and Silva, S. J.: Importance of Dry Deposition Parameterization Choice in Global Simulations of Surface Ozone., Atmospheric Chemistry & Physics Discussions, 4, https://doi.org/10.5194/acp-2019-429, 2019.

Wu, Z., Staebler, R., Vet, R., and Zhang, L.: Dry deposition of O3 and SO2 estimated from gradient measurements above a temperate mixed forest, Environmental pollution, 210, 202–210, https://doi.org/10.1016/j.envpol.2015.11.052, 2016.

Wu, Z., Schwede, D. B., Vet, R., Walker, J. T., Shaw, M., Staebler, R., and Zhang, L.: Evaluation and intercomparison of five North American dry deposition algorithms at a mixed forest site, Journal of Advances in Modeling Earth Systems, 10, 1571–1586, https://doi.org/10.1029/2017MS001231, 2018.

Young, P. J., Naik, V., Fiore, A. M., Gaudel, A., Guo, J., Lin, M., Neu, J., Parrish, D., Rieder, H., Schnell, J., et al.: Tropospheric Ozone Assessment Report: Assessment of global-scale model performance for global and regional ozone distributions, variability, and trends, Elem Sci Anth, 6, https://doi.org/10.1525/elementa.265, 2018.

Zhang, L., Brook, J. R., and Vet, R.: On ozone dry deposition–with emphasis on non-stomatal uptake and wet canopies, Atmospheric Environment, 36, 4787–4799, 2002.

Zhang, L., Brook, J., and Vet, R.: A revised parameterization for gaseous dry deposition in air-quality models, Atmospheric Chemistry and Physics, 3, 2067–2082, 2003.