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Development and technical paper | 04 Sep 2015

A new chemistry option in WRF-Chem v. 3.4 for the simulation of direct and indirect aerosol effects using VBS: evaluation against IMPACT-EUCAARI data

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Received: 08 Dec 2014 – Discussion started: 03 Feb 2015 – Revised: 23 Jul 2015 – Accepted: 31 Jul 2015 – Published: 04 Sep 2015

Abstract. A parameterization for secondary organic aerosol (SOA) production based on the volatility basis set (VBS) approach has been coupled with microphysics and radiative schemes in the Weather Research and Forecasting model with Chemistry (WRF-Chem) model. The new chemistry option called "RACM-MADE-VBS-AQCHEM" was evaluated on a cloud resolving scale against ground-based and aircraft measurements collected during the IMPACT-EUCAARI (Intensive Cloud Aerosol Measurement Campaign – European Integrated project on Aerosol Cloud Climate and Air quality interaction) campaign, and complemented with satellite data from MODIS. The day-to-day variability and the diurnal cycle of ozone (O_3) and nitrogen oxides (NO_x) at the surface are captured by the model. Surface aerosol mass concentrations of sulfate (SO_4), nitrate (NO_3), ammonium (NH_4), and organic matter (OM) are simulated with correlations larger than 0.55. WRF-Chem captures the vertical profile of the aerosol mass concentration in both the planetary boundary layer (PBL) and free troposphere (FT) as a function of the synoptic condition, but the model does not capture the full range of the measured concentrations. Predicted OM concentration is at the lower end of the observed mass concentrations. The bias may be attributable to the missing aqueous chemistry processes of organic compounds and to uncertainties in meteorological fields. A key role could be played by assumptions on the VBS approach such as the SOA formation pathways, oxidation rate, and dry deposition velocity of organic condensable vapours. Another source of error in simulating SOA is the uncertainties in the anthropogenic emissions of primary organic carbon. Aerosol particle number concentration (condensation nuclei, CN) is overestimated by a factor of 1.4 and 1.7 within the PBL and FT, respectively. Model bias is most likely attributable to the uncertainties of primary particle emissions (mostly in the PBL) and to the nucleation rate. Simulated cloud condensation nuclei (CCN) are also overestimated, but the bias is more contained with respect to that of CN. The CCN efficiency, which is a characterization of the ability of aerosol particles to nucleate cloud droplets, is underestimated by a factor of 1.5 and 3.8 in the PBL and FT, respectively. The comparison with MODIS data shows that the model overestimates the aerosol optical thickness (AOT). The domain averages (for 1 day) are 0.38 ± 0.12 and 0.42 ± 0.10 for MODIS and WRF-Chem data, respectively. The droplet effective radius (R_e) in liquid-phase clouds is underestimated by a factor of 1.5; the cloud liquid water path (LWP) is overestimated by a factor of 1.1–1.6. The consequence is the overestimation of average liquid cloud optical thickness (COT) from a few percent up to 42 %. The predicted cloud water path (CWP) in all phases displays a bias in the range +41–80 %, whereas the bias of COT is about 15 %. In sensitivity tests where we excluded SOA, the skills of the model in reproducing the observed patterns and average values of the microphysical and optical properties of liquid and all phase clouds decreases. Moreover, the run without SOA (NOSO) shows convective clouds with an enhanced content of liquid and frozen hydrometers, and stronger updrafts and downdrafts. Considering that the previous version of WRF-Chem coupled with a modal aerosol module predicted very low SOA content (secondary organic aerosol model (SORGAM) mechanism) the new proposed option may lead to a better characterization of aerosol–cloud feedbacks.

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How to cite: Tuccella, P., Curci, G., Grell, G. A., Visconti, G., Crumeyrolle, S., Schwarzenboeck, A., and Mensah, A. A.: A new chemistry option in WRF-Chem v. 3.4 for the simulation of direct and indirect aerosol effects using VBS: evaluation against IMPACT-EUCAARI data, *Geosci. Model Dev.*, 8, 2749–2776, <https://doi.org/10.5194/gmd-8-2749-2015>, 2015.

