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A mitigation study for air pollution management across Germany for NO_x (NO + NO₂) with the LOTOS-EUROS CTM – Part I: Comparing the labeling and brute force technique for source attribution.

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Air quality is a key aspect of present environmental discussions with nitrogen oxides (NO_x = NO + NO₂) has become a decisive element and impact factor for air quality planning. Millions of people are exposed by NO₂, especially in urban areas near traffic sites, leading to increased mortality rates. As the annual limit value of 40 µg/m³, introduced by the European Ambient Air Quality Directive (EC, 2008), is currently exceeded by about 39 % (UBA, 2019), in Germany an estimated number of 13.100 premature deaths are caused by NO₂ (EEA, 2018). The origin and formation processes of NO_x are well documented in literature for long: NO mainly originates from incomplete combustion (Granier et al., 2011; Vestreng et al., 2009), with NO₂ formed as a photochemical reaction product (Finlayson-Pitts and Pitts, 2000; Leighton, 1961). Therefore, to further improve the ambient air quality using cost-effective mitigation strategies, this requires for quantifying the contribution of the ambient air pollution by source sectors and regions of their origin (Belis et al., 2020).

Applying chemical transport models (CTMs) for source attribution (SA), one can distinguish between contributions and impacts. Methods to estimate contributions are known as labeling (Kranenburg et al., 2013) or tagging (Wang et al., 2009; Wagstrom et al., 2008) approaches and are based on conservation of mass. In contrast, sensitivity simulations, such as the top-down brute force (BF) technique, can be used to quantify the impact to different emission reductions (Clappier et al., 2017; Thunis et al., 2019). As the BF approach in theory is only designed for impact studies, the calculation of contributions can result in incorrect estimates which is dependent on the linearity of the considered component (Clappier et al., 2017; Thunis et al., 2019). Therefore, impact studies can only be employed under certain restrictions and their application range needs to be predefined first (Thunis et al., 2020).

Previous studies primarily focused on PM when comparing different approaches for SA. Therefore, we conducted a SA study by performing air pollution simulations using the LOTOS-EUROS CTM across Germany of January 1st to December 31st, 2018 for NO_x. We enhanced the understanding of limitations to non-linear interaction terms and defined the potential application range for SA purposes using impact studies of NO_x, by comparing the labeling approach implemented in the LOTOS-EUROS CTM to the BF technique.

First results indicate that impact studies cannot be used to estimate contributions of NO due to their non-linear relations and inconsistent mass conservation. Even though differences for NO₂ are smaller, it is not recommended to apply the BF technique here either. However, considering that non-emission sources cannot be separated from each other in impact studies, it is further advised not to apply this method for NO_x.