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Ozone time scale decomposition and trend assessment from surface observations

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Emissions of ozone precursors have been regulated in Europe since around 1990 with control measures primarily targeting to industries and traffic. In order to understand how these measures have affected air quality, it is now important to investigate concentrations of tropospheric ozone in different types of environments, based on their NO_x burden, and in different geographic regions. In this study, we analyze high quality data sets for Switzerland (NABEL network) and whole Europe (AirBase) for the last 25 years to calculate long-term trends of ozone concentrations. A sophisticated time scale decomposition method, called the Ensemble Empirical Mode Decomposition (EEMD) (Huang,1998;Wu,2009), is used for decomposition of the different time scales of the variation of ozone, namely the long-term trend, seasonal and short-term variability. This allows subtraction of the seasonal pattern of ozone from the observations and estimation of long-term changes of ozone concentrations with lower uncertainty ranges compared to typical methodologies used. We observe that, despite the implementation of regulations, for most of the measurement sites ozone daily mean values have been increasing until around mid-2000s. Afterwards, we observe a decline or a leveling off in the concentrations; certainly a late effect of limitations in ozone precursor emissions. On the other hand, the peak ozone concentrations have been decreasing for almost all regions. The evolution in the trend exhibits some differences between the different types of measurement. In addition, ozone is known to be strongly affected by meteorology. In the applied approach, some of the meteorological effects are already captured by the seasonal signal and already removed in the de-seasonalized ozone time series. For adjustment of the influence of meteorology on the higher frequency ozone variation, a statistical approach based on Generalized Additive Models (GAM) (Hastie,1990;Wood,2006), which corrects for meteorological effects, has been developed in order to a) investigate if trends are masked by meteorological variability and b) to understand which part of the observed trends is meteorology driven. By correlating short-term variation of ozone, as obtained from the EEMD, with the corresponding short-term variation of relevant meteorological parameters, we subtract the variation of ozone concentrations that is related to the meteorological effects explained by the GAM. We find that higher frequency meteorological correction reduces further the uncertainty in trend estimation by a small factor. In addition, the seasonal variability of ozone as obtained from the EEMD has been studied in more detail for possible changes in its behavior. A shortening of the seasonal cycle was observed, i.e. reduction of maximum and in-crease of minimum concentration per year, while the occurrence of maximum is shifted to earlier times during a year. In summary, we present a sophisticated and consistent approach for detecting and categorizing trends and meteorological influences on ozone concentrations in long-term measurements across Europe.