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Impact of the transport of aerosols from the free troposphere towards the boundary layer on the air quality in the Paris area

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Abstract

We propose a quantification of the downward transport of aerosols from the free troposphere (FT) to the planetary boundary layer (PBL). Aerosols are originally released at the surface as a consequence of anthropogenic activities, biomass burning, soil mobilization, etc. After being vertically transported into the FT, they are exposed to the long-range transport (LRT) and may subside to impact, in turn, surface air pollution in remote places. Using 5400 h of routine Lidar observations conducted at the SIRTA observatory in the Paris area (France), we identified 154 free tropospheric aerosol layers continuously monitored during their downward transport into the local PBL. One of these events—associated to a Saharan dust outbreak—is thoroughly documented in a case study. And a climatological analysis of surface PM10 levels recorded at air quality monitoring stations allows the impact of FT to PBL transport of aerosols to be quantified. This source is found to be significant for 15 out of the 16 stations, with average PM10 concentrations 2.14 $\mu g\,m^{-3}$ (i.e. 12%) above climatological values after the injection of free tropospheric aerosols into the PBL.

Keywords: Aerosols; Air quality; Long-range transport; Free troposphere–boundary layer coupling; Lidar

1. Introduction

The understanding of the role that atmospheric particulate matter plays in the earth system is a subject of growing concern. Partly because our level of understanding of the processes involved in their formation, transport, and transformation is low, but also because they have adverse impacts on the environmental equilibrium of the planet—including their role in the climate forcing (direct and indirect radiative impacts, IPCC, 2001), and their impact on air quality and subsequently on human health (e.g. Moshammer and Neuberger, 2003).

This study focuses on the factors of variability of aerosols in the planetary boundary layer (PBL) in an urbanized area of Western Europe; hence, the primary motivation regards air quality. In such areas, anthropogenic emissions of aerosols and precursors of secondary organic aerosols (SOAs) are thought to be a major contributor to the budget of PM10 (particulate matter $<10\,\mu m$ in equivalent diameter). But the respective role of anthropogenic emissions, soil mobilization, and import from distant places is very uncertain (Bessagnet et al., 2005). In the context where the European legislation (EU Directive 1999/30/CE) requires a 50% decrease

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of annual mean PM10 levels between 2005 and 2010 (from 40 to 20 $\mu g\,m^{-3}$), more precise partitioning of each contribution is needed in order to design emission reduction strategies.

The main sources of aerosols in the Western European atmosphere are Saharan mineral dusts (Dulac et al., 1992; Moulin et al., 1998), resuspension of local terrigeneous matter (Vautard et al., 2005), anthropogenic SOA (Wandinger et al., 2002), biomass burning SOA (Real et al., 2007), and volcanic SOA (Chazette et al., 1995).

The transport cycle of aerosols can be split in three parts. Firstly, primary aerosols and precursors of SOA are emitted at the surface; consequently they primarily affect PM10 levels in the boundary layer. Secondly, aerosols may be transported towards the free troposphere (FT) where they play a role on the variability of trace species and are exposed to the long-range transport (LRT) (e.g. Stohl et al., 2002a). Thirdly, after their transport in the FT, they may be transported downwards back into the PBL and contribute to surface pollution levels at distant places.

The quantification of the downward transport of aerosols remains challenging because of the dynamical barrier constituted by the top of the PBL. The most widespread approach consists in documenting the fate of a layer observed in the FT by means of Lagrangian modeling and to correlate an increase of surface levels to the predicted trajectory of the air mass (e.g. Gerasopoulos et al., 2006a; Real et al., 2007). However, considering the processes involved, modeling the injection of an air mass from the FT into the PBL requires the use of a mesoscale model. Consequently, such investigations remain limited to case studies, and cannot be integrated to provide a global estimate of the contribution of LRT to surface PM10 levels.

Alternative methods consist in taking advantage of additional measurements in the PBL. By monitoring dust optical thickness in the PBL, Hamonou et al. (1999) could identify an isolated case of Saharan dust transport to the European PBL. The high Lidar depolarization ratio of Saharan dust allowed Gobbi et al. (2007) to quantify its impact on surface air quality in Italy. The composition of particulate matter can also be used as a proxy to source apportionment (e.g. Querol et al., 2001). A similar procedure led Rodríguez et al. (2002) and Gerasopoulos et al. (2006b) to show the significance of the contribution of Saharan dust to PM10 levels in Spain and Greece, respectively, but the importance of this source on air quality in France is uncertain.

Here, we propose a new approach to estimate the contribution of downward transport from the FT to the PBL to particulate air pollution in the Paris area. Trace species are transported over long distances in the FT in layers (Newell et al., 1999; Hamonou et al., 1999) before being mixed with the background (Colette and Ancellet, 2006). Lidars have proved to be a key instrument in the documentation of the characteristics of free tropospheric layers (Stohl and Trickl, 1999; Matthias et al., 2004; Ravetta et al., 2007). The continuous temporal coverage that they offer allows tropospheric layers to be monitored during their downward transport until they reach the PBL (Tsunematsu et al., 2006). A Lidar designed for aerosol and cloud studies has been routinely operated since 2002 in the Paris area. This large dataset of Lidar profiles makes the investigation of a significant number of events possible.

The dataset of Lidar profiles is presented in Section 2 together with a climatological analysis of the variability of aerosol layers in the FT. A case study of LRT of aerosols followed by an injection into the PBL is discussed in Section 3 to illustrate the processes considered in Section 4 that addresses in details the quantification of the overall impact of downward transport of aerosols on surface air quality.

2. Database of aerosol Lidar profiles

2.1. The SIRTA observatory

Aerosol profiles used in this study were collected at the Site Instrumental de Recherche par Télédétection Atmosphérique (SIRTA; Haeffelin et al., 2005). The SIRTA is an observatory created in 1999 by Institut Pierre-Simon Laplace and Ecole Polytechnique. The site is involved in several observation networks such as Earlinet (Bösenberg et al., 2003). It is located 20 km southwest of Paris in a suburban environment occasionally exposed to pollution plumes of the Paris city. The Paris area is ideally located for the investigation of LRT because of its position at the crossroad between major pathways of pollution transport (Stohl et al., 2002a).