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Evaluation of the parametrized transport of lead-210 in high-altitude European sites

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Abstract

One important aspect in current climate modelling is to correctly simulate the regional variability of aerosols forcings. A first step to address this issue is to evaluate the tropospheric vertical distribution of submicronic aerosol (including sulfate and organic aerosol) in particular in the free troposphere.

^{210}Pb is particularly interesting to study the dynamic transport by chemistry and transport models, because it is an excellent passive tracer of atmospheric circulation. Hence, it is a simple representative of atmospheric submicronic aerosol and its transport is well correlated with sulfate aerosol. Indeed, ^{210}Pb and ^{222}Rn (its radioactive parent) have been widely used for model verification. We present here pluriannual simulations of ^{210}Pb made with the multiscale global to regional model MOCAGE of Météo-France, which allows to investigate resolution effects over Europe and specially anomalies in link with the heat wave episode of summer 2003 and associated increased convection.

Precisely, simulations over Europe over 3 years, 2002 to 2004, are compared with the original dataset gathered in particular over high-altitude sites during the CARBOSOL project. We particularly focus our attention on the vertical unresolved-scale transport parameterisation and analyse the strong seasonality in ^{210}Pb concentrations (maximum in summer, minimum in winter), which was exceptionnally marked in 2003.

1 Introduction

One of the challenges in climate modelling is to correctly simulate the regional aerosols forcings on the Earth radiative budget (IPCC Working Group, 2008). One reason for the difficulties in estimating the radiative forcings by aerosols is the important spatial and temporal variabilities in concentrations. Indeed aerosols have a short lifetime in the atmosphere, typically between some days and some weeks, resulting from the different source and sink terms for each aerosol specie (Charlson et al., 1992). For these

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reasons, a first step to adress this issue is to evaluate the tropospheric vertical and horizontal distribution of submicronic aerosol (including sulfate and organic aerosol) at sufficiently fine resolutions.

Sulfate aerosol absorbs solar radiation and is a condensation nuclei (Langner et al., 1992). Thus, it has a negative radiative forcing, that counters at the regional to continental scales the positive forcings from other aerosol (black carbon,...) and from greenhouse gases. It is indeed now an important focus in global Chemistry-Climate Models (CCM). However, due to the short lifetime of sulfate aerosol in the atmosphere (Charlson et al., 1992), it is essential to simulate accurately the distribution at the regional to continental scales and in the vertical. Modeling sulfate in a CCM or Chemistry-Transport Model (CTM) is however associated with several uncertainties, from the emissions of gas phase precursors to the chemical reactions and kinetics in both aqueous and gas phase, including also effects linked with the limited horizontal and vertical resolutions that are tractable with current high-performance computational equipment. Eventhough sulfate model evaluation can be directly made using sulfate observation, it is needed to separate the evaluation of the representation of transport (both at resolved and unresolved scales) and wet deposition, processes which strongly impact on sulfate aerosol distributions, and the one of emissions and chemical processes, as the effects can indeed average out.

^{210}Pb is a representative of atmospheric submicronic aerosols. Observations of ^{210}Pb indicate a strong correlation with sulfate aerosol (Hammer et al., 2007), as illustrated for instance on Fig. 1 at the Sonnblick site, and ^{210}Pb is an excellent tracer of processes of transport at resolved and sub-grid scales and of wet scavenging (its main atmospheric sink) in the atmosphere. Thus, many studies have used concentrations of ^{210}Pb in order to evaluate the representation of scavenging and/or dry deposition processes in models (Balkanski et al., 1993; Rehfeld and Heimann, 1995).

Some authors (Lee and Feichter, 1995; Giannokopoulos et al., 1999) performed intercomparisons between several advection and parameterisation schemes in order to evaluate the representation of transport and wet deposition processes in their CTM or

CCM. However, most of these studies have been carried out at the global scale, with low vertical and horizontal resolutions. Generally, the observational data used are climatologies in remote areas, with few points actually in Europe. In the present study, after evaluating our model at the global scale using this approach, we focus on the evaluation of regional transport up to European high-altitude sites using the original CARBOSOL dataset and by refining our model horizontal resolution over the area of interest.

2 Model description and evaluation at the global scale

2.1 The chemistry and transport model MOCAGE

MOCAGE (Peuch et al., 1999) (Atmospheric Model of Chemistry on a Large Scale) is the three-dimensional chemistry and transport model of Météo-France. It is an off-line model based on a semi-lagrangian scheme of advection (Josse et al., 2004). On the vertical, the model has 47 levels from the ground to 5 hPa in hybrid (sigma, Pressure) coordinates, making it possible for the vertical levels to be terrain-following near the surface while being isobars in the stratosphere. This vertical extension allows to account for transport and chemical processes from the planetary boudary layer, to the free troposphere stratosphere. MOCAGE is also a multi-scale model: it is possible to use several two-ways nested domains over areas of interest. In this study, we have used two configurations. The first one (Sect. 2.2) has a global domain with a 2° resolution and a European domain with a 1° resolution. The second configuration used (Sect. 3.4) has a coarser global domain (4° horizontal resolution), but a larger and finer European grid at 0.5° resolution. The choice of model configuration and geometry is flexible for the user, depending on application: climate-chemistry (Teyssedre et al., 2007), air quality (Dufour et al., 2004; Honore et al., 2008; Hollingsworth et al., 2008), long-range transport of polluants or chemical data assimilation (Bousserez et al., 2007; Pradier et al., 2006; Clark et al., 2007; Barret et al., 2008).

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Atmospheric forcings needed by the CTM (pressure, temperature, horizontal wind and relative humidity) are meteorological analyses from the operation numerical weather prediction suite of Météo-France ARPEGE (Courtier and Geleyn, 1988). One of the characteristics of MOCAGE is its modular concept. It comprises a comprehensive package of optional physical and chemical parameterisations, that can be chosen by the user. For the present study, only 2 tracers are considered: radon and lead.

The emission rate of ^{222}Rn is a very important input to the simulations. The commonly used emission rate is Eo=1 atom.cm^{-2}.s^{-1} above non-frozen areas and 0 above the permafrost areas and oceans (Jacob et al., 1997). It has however been put into question in recent years, specially for high latitudes. We have thus chosen the approach proposed by (Schery, 2004), maintaining this flux value of Eo up to 30° N only, but then decreasing linearly down to 0.2 atom.cm^{-2}.s^{-1} at 70° N, and nullifying above 70° N. This estimate for the ^{222}Rn emissions flux is in agreement with observations of (Conen and Roberston, 2002), which showed evidence of a North-South gradient of Radon emissions.

Dry deposition has been parameterized simply, following (Guelle et al., 1998): deposition velocity is 0 on ice, 0.012 cm.s^{-1} on water and 0.12 cm.s^{-1} on the ground. Following also (Guelle et al., 1998) and the majority of other similar works, sedimentation has been neglected, due to the small size of aerosols carrying ^{210}Pb.

Convection and the turbulent diffusion are essential sub-gridscale transport and mixing processes. Preunkert and Wagenbach (1998) consider in particular that turbulence and convection are primarily responsible for the transport of the pollutants from the low atmospheric layers to the high troposphere, as sampled for instance in high altitude sites. It is thus of importance that these two processes are correctly parametrised in the MOCAGE model. The parametrisations schemes used for these two phenomena are described in detail and have been evaluated by Josse et al. (2004). Their description is summarized here. For turbulent diffusion, the scheme of Louis (Louis, 1979) has been introduced into MOCAGE. The parameterisation of convection used in this study is the mass flux approach of Bechtold et al. (2001). This scheme is of particular

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interest for us, as ice and snow are accounted for. Moreover, whashout processes are also directly implemented within the convective scheme, following Mari et al. (2000). Stratiform rainout uses the parameterization of Liu et al. (2001), with a modification for below-cloud scavenging. We consider that the washout is done by snowflakes between 233 K and 268 K and by rain drops for temperatures higher than 268 K.

2.2 Evaluation at the global scale

At the global scale, ^{210}Pb simulations of MOCAGE are evaluated here on the basis of two one-year runs, covering 2001 and 2002. For every yearly simulation, a spin-up period of 1 month is enough to be on a steady state in the free troposphere, starting from zero-concentrations, ^{210}Pb coming only from the radio-active decay of ^{222}Rn, which has a lifetime of approximately 5 days.

Table 1 describes the geographical coordinates of the different observational sites considered in our study. Observations have been obtained from the database of Preiss et al. (2000), except for Hachijojima, Okushiri et Chichijima, which were taken from Feichter et al. (1991).

For every station, simulated concentrations have been linearly interpolated on the horizontal at the exact geographical location of the station, and taken at the lowest level of the model. However, for Izania and Mauna Loa, both situated on relatively small islands, MOCAGE global resolution (2°) cannot match the high altitude of these points. The comparison with observations for these two sites is made by considering modelled concentrations at the model pressure level corresponding to their actual altitude (standard atmosphere). The results are presented on Fig. 2.

At Northern high latitudes (Fig. 2a), the annual cycle of ^{210}Pb is well reproduced, in particular for the winter maximum. However, the magnitude of the cycle is too low, as already diagnosed for other modelling results by Rehfeld and Heimann (1995), Guelle et al. (1998) or Giannokopoulos et al. (1999). Numerous factors can explain this typical behaviour, in particular possibly the current crudeness of the scheme accounting for deposition due to ice and snow. At Kap Tobin, the november maximum is not repro-

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duced. However, observations are based on 2 years of observations only, and are may be not representative as a climatology. The annual intervariability may indeed be high, even in model simulations, as illustrated by the differences between 2001 and 2002 MOCAGE winter concentrations at Point Barrow.

For North American stations, the annual cycle is well reproduced, and mean biases are small. For New York, the model is particularly close to the climatology. However, for Beaverton the behaviour of the model is similar to the one for high latitude sites: results are fair but the magnitude of the annual cycle is underestimated. In Asia, the impact of the moonsoon, which drives the annual cycle, is very well represented. At Okushiri, results are quite good, whereas at Hachijojima, high values are over-estimated with a factor 2.

The observed annual variations at Bermuda Island are quite low, in agreement with the observed precipitations (not shown). However, the simulated cycle of precipitations is pronounced, with strong precipitations in summer, which can explain the minimum of simulated ^{210}Pb concentrations.

Over Europe, annual cycles are well reproduced, with a strong interannual variability at Orsay. At Mace Head, the low magnitude of the cycle is due to the majoritary influence of atlantic air masses, as detailed for radon in Josse et al. (2004).

For tropical and subtropical latitudes (Fig. 2c), simulated concentrations are very close to the observations, emphasizing that the representation of convective rainfall in the model is sufficient for the purpose of representing the scavenging of soluble species. Only Miami and Guayaquil are exceptions, with very poor results. For Guayaquil, the cycle is in phase with observations, but the summer concentrations are much too high. However, this has not to be linked to rainfall, since at this period of the year, climatological precipitations are extremely low (1 mm per month), and are actually slighty over-estimated in the model. The discrepancy between model and observations is rather due to wrong vertical mass fluxes at this partiular point. Guyaquil is indeed located downwind from the strong orography of the Andes. The hypothesis of hydrostatism show probably here its limits. For Miami, MOCAGE winter concentrations are

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too high compared to observations, and the annual cycle magnitude is also too high, especially for 2002. A possible explanation is the influence of local sea breezes that lead to mixing with oceanic ^{210}Pb levels, which have less intra-annual variability. The specific shape of the tip of Florida and the scale of these breezes are probably too small to be seen with a resolution of 2 degrees.

Concerning sites in the mid- and high latitudes of the Southern Hemisphere (Fig. 2d) and far from the radon sources (G Von Neumayer, Invercargill and Chatham), the results are fair: ground concentrations are in the observed range. However, over Antarctica, the annual cycle is not well reproduced, following what has been investigated for radon in Josse et al. (2004). Closer to the sources, the annual cycle is good, and biases are low. At Norfolk, a strong interannual variability appears in May–June, and 2002 appears to be far from the climatology.

To conclude this section, MOCAGE is able to reproduce annual cycles and average concentrations of ^{210}Pb at the global scale except for only one station (Guayaquil), where local phenomena in link with a very marked orography probably prevail. The behaviour of the model seems particularly good at tropical latitudes, where uncertainties on radon emission rates are the lowest.

3 Evaluation at the European scale

3.1 The CARBOSOL dataset

As already mentioned, the purpose of the ^{210}Pb simulations presented in this paper was to evaluate the capacity of MOCAGE to transport pollutants from the boundary layer to high troposphere. In fact, ^{210}Pb and aerosol sulfate are transported, washed in the same way. They are a valuable first step in the evaluation of the model representation of the sulfur cycle over Europe.

For this work, we used measurements of the CARBOSOL project (Legrand and Puxbaum, 2007). The CARBOSOL project (Present and Retrospective State of Or-

ganic versus Inorganic Aerosol over Europe: Implications for Climate, finished in 2005) combined the study of spatial distribution and composition of the present-day carbonaceous aerosol in air and wet deposition in six stations in Europe with the composition trends of climatically relevant carbonaceous material in Alpine ice cores spanning pre-industrial period. The observational sites were instrumented in order to collect aerosols in areas geographically and climatologically different. Their detailed description can be found in Pio et al. (2007) and in Hammer et al. (2007). In our study, we did not use Azores data, as we decided to focus on main land Europe with a special focus on altitude sites. So, we had only five stations of measurements for our comparison; they are described in Table 2.

For these stations, the filters were changed in general once a week. Their analysis by the various European partners of CARBOSOL project made possible to obtain data sets of the organic, mineral aerosols (sulfate in particular), but also ^{210}Pb by Hammer et al. (2007).

3.2 Comparison of ^{210}Pb simulations and observations in altitude sites

Altitude sites provide very interesting insight on background concentrations as they are generally not under the direct influence of emission sources. Marenco et al. (1994) have for instance use historical data at several sites to derive decenal trends in tropospheric ozone over Europe. However, comparing the results of numerical model in such montaineous and altitude sites is a challenge as, firstly, the horizontal resolution of models generally do not allow for an accurate representation of the terrain (for instance, with our 1° domain, the Alps peak at approximately 2500 m while the Sonnblick site is at 3100 m, as described in Table 2) and, secondly, dynamic processes in or near the orography cannot be accounted for with the non-hydrostatic formulation that is generally used in large-scale models. Also, non-hydrostatic models that can account well of such processes (Bougeault et al., 2001) cannot be run over long periods of time (i.e. several months), which are needed to study seasonal and interannual variability.

An objective of our work was to investigate ^{210}Pb simulations in the altitude sites

of CARBOSOL. The zoom option of MOCAGE has been used to compare resolution effects, both direct and indirect (through application of parameterisation at increased resolution, that better accomodates gradients,...).

After different tests, we decided to compare the observed data with the simulations at the model level of the pressure corresponding to the altitude of the site (in standard atmosphere), often located in the free atmophere for the model. This method, that we have already used for the evaluation at the global scale (Sect. 2.2), appears to be best able to capture the characteristics of the observed variability in the high altitude sites. This choice in fact implies that there is little or no dry deposition of the aerosols under study, hypothesis confirmed by M. Legrand, personal communication: in Europe, it rains regulary and dry deposition is insignificant in comparison with wet scavenging.

3.3 Simulations of 2002, 2003 and 2004

Simulations were made with the three domain configuration described in Sect. 2.1.

Firstly, we calculated the annual average and compared with measurements of Hammer et al. (2007) (see Table 3). The annual averages simulated by MOCAGE are close to the observed ones. This result confirms in particular the validity of the latitudinal gradient for the Radon emissions that we have introduced.

The seasonal averages also presented in Table 3 indicate that the annual cycle is well represented for sites in plains and the simulated data are generally in close agreement with measurements (see Fig. 3). A systematic bias (slightly too high values in winter and slightly too low values in summer) is however observed indicating that the model presents a somewhat reduced amplitude for the annual cycle. In winter, the overestimation of ^{210}Pb concentrations may have two origins: it could be that vertical stratification is too strong in the model, in link with the coarse vertical resolution (at most 7 levels in the planetary boundary layer) and the crudeness of the turbulence scheme of Louis, or that Radon emissions are overestimated with an unappropriate representation of the occurence of frozen soil. In summer, the model underestimates the Lead concentration, especially for high altitude sites. One error source could be

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the representation of transport by convection, as studied hereafter.

At a finer time scale, the annual evolution (simulated and measured) of the contents of ^{210}Pb is presented for each CARBOSOL station on Fig. 4. The results are plotted using the timesteps of measurements (about one week). For Aveiro, MOCAGE reproduces well the strong variability of the ^{210}Pb concentrations. Aveiro is located on the Portuguese coast; relying on meteorological analyses, the CTM captures well the origin (land or sea) of the air masses that are sampled, that have very contrasted ^{210}Pb contents. For the second station in low altitude K-puzta, located in the Hungarian plain, the presence of the anticyclone of Siberia implies a stagnation of the aerosols in boundary layer, during the winter. This phenomenon is well reproduced by the model. It is in phase with strong ^{210}Pb concentrations of measurements. However, it seems to underestimate the summer concentrations of 2003.

The analysis of the results obtained for the 3 stations of middle and high altitude show that the 3 annual cycles are well reproduced. However, a specificity appears for the summer 2003 with a marked underestimation of our simulation. In the station of the Austrian Alps, Sonnblick, there is also a factor 2 between observations and simulations. This is of strong interest since the summer of 2003 was characterized by an exceptional heat wave across Europe and particularly in France (Levinson and Waple, 2004). The heat wave had consequences on the ^{210}Pb distribution, which the model apparently cannot capture as well for this year as for 2002 or 2004. Yet the simulated concentrations correctly increase during winter and spring 2003; for the end of May 2003 to the beginning of September 2003, they are abnormally low in comparison with observations.

3.4 Spring and summer 2003

3.4.1 Meteorological conditions during spring and summer 2003

We have chosen to illustrate the meteorological conditions of the spring and summer of 2003 using four specific days (see the geopotential maps at 500 hPa on Fig. 5). We

give first a general overview of the period and of its specificities.

In April, the weather was nice: temperatures increased gradually and maxima reached already 30°C at the end of the month. Precipitations were lower than World Meteorological Organisation monthly normals. The first days of May were also warmer than the normals, both for the minimal and maximal temperatures and all across Europe. The weather was a bit unstable but dry. Starting from mid-May, it became wet, more stormy, with temperatures close to monthly normals. This was due to the crossing of several perturbed weather systems.

At beginning of June daily maximum temperatures increased markedly in particular close to the Alps. Only Scandinavia was not concerned by this increase. The precipitations were scarce and the weather was sometimes stormy. The larger part of Europe was under very weak barometric gradients. Atlantic fronts were rejected towards Northern Europe. The weather was unstable and kept stormy until the middle of June. After, a high geopotential settled over the Azores. His ridge influenced all Western Europe.

In July, temperatures were close to normals. Precipitations were mainly due to stormy showers. In the first two weeks of August, a heat wave struck most of European countries (Levinson and Waple, 2004), except in the North (Scandinavia) and in the South (Greece and Romania). The deviations of mean temperatures from normals reached 4°C. At 500 hPa, a ridge spread from Gibraltar to the Netherlands (Bessemoulin et al., 2004). The air mass was stable and large-scale convection was inhibited. But, close to orography, daily thunderstroms occured: they were generally dry, but, sometimes, violent with precipitations of hail. On the 13th of August, seven paragliding pilots took of from Chamonix (elevation 1000 m) and succeeded in landing on the Mont-Blanc top (4807 m): huge upslope winds took place close to the mountains.

We consider four different days in the following: a hot and dry day of spring (May 3rd), the 22nd of May during a convective precipitations period, the 9th of June with high temperatures, thunder but little precipitations, and finally, a very hot day during the second part of the heat wave (4th August).

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3.4.2 ^{210}Pb simulation

We investigate here the lower than observed modelled concentrations during the spring and summer 2003 in high-altitude CARBOSOL sites. The modelled ^{210}Pb concentrations at 700 hPa (model pressure level close to Sonnblick's altitude) are reproduced on Fig. 6.

In order to investigate the sensitivity of simulations to the horizontal resolution of the CTM, we have run MOCAGE with another configuration (see Sect. 2.1), that has a better resolution over Europen (0.5° instead of 1°). ^{210}Pb concentrations obtained with this second configuration are also plotted on the 700 hPa isobar on Fig. 7.

The simulated concentrations are similar for both resolutions for the 3rd and the 22nd of May 2003. As mentionned in Sect. 3.2, ^{210}Pb concentrations were indeed satisfactory for the five sites and for these two days, even at 1° resolution. At variance, ^{210}Pb concentrations at 700 hPa are significantly higher with the 0.5° resolution than with the 1° resolution for the other days (9th of June, and very strinkingly for 4th August). In some places, the difference between the two simulations reaches 50%. As the 1° were much underestimated compared to observation, the increase in model horizontal resolution brings an improvement for high altitude ^{210}Pb levels during the heat wave. Figure 8 investigates the origin of such differences, by presenting a vertical cross-section of both ^{210}Pb and ^{222}Rn for the 0.5° resolution, at the latitude of the Sonnblick site (47° N) and for 4th August) at 12:00 UTC. The mixing ratios of ^{210}Pb are quite homogeneous (except for a vertical column around 7° E corresponding to a "fresh" convective event with some precipitations in the model), with values of around 3.10^{-20} mol.mol^{-1} from the surface up to model level 16–17, corresponding to an average altitude of 5000 m above sea level. All the sites of CARBOSOL are within this thick layer, explaining the unusually high values sampled in this period. If one considers now ^{222}Rn (bottom pannel), one sees that it is only distributed at this date over 5 or 6 model levels (2000 m approximately) corresponding broadly to the height of the planetary boundary layer, except for the convective mixing area already mentioned at 7° E. This

indicates that the situation for ^{210}Pb is not the result of "fresh" Radon desintegration, but rather is a continental/seasonal feature, which is the result of several past weeks of vertical transport without significant precipitating events, the main atmospheric sink for ^{210}Pb.

The simulated concentrations for the both resolutions, 0.5° and 1°, are compared with the observed ones (for the 5 validation sites, see Sect. 3.1) for the spring and summer 2003 (Fig. 9). Just before the convective and rainy period at the beginning of May 2003, MOCAGE was very close to the observations for the two configurations. From 12th to 26th of May, the scavenging was high because there was convective and thundery precipitations. Concentrations in observations and in the model were both low.

Then, at the beginning of June, the conditions become hotter and more unstable. The observed ^{210}Pb concentrations increased both in mountainous sites and in plain sites. Quite the opposite, the concentrations simulated by MOCAGE stay low in spite of this weather change, whatever the resolution (0.5° or 1°). Aveiro is an exception, probably because its location under oceanic influence. After the middle of June, the high geopotential over Western Europe prevented precipitations and washout. The simulated concentrations increased only for the 0.5° resolution and were close to observations during July. At beginning of August, during the second part of the heat wave, the simulated high concentrations of ^{210}Pb were in phase with the maxima of pressure, but with values lower than observated concentrations. All the same, one notices that the 0.5° resolution results are better, they cannot reach values as high as in the observations. The interpretation of differences between the 0.5° and 1° simulations is principally linked to the parameterisation of turbulence and convection: in the 0.5° one, subgrid-scale parameterized transport is applied over model gridcells that are four time smaller than at 1°; it may well be that only one or two of the 4 cells have a turbulent and convective character (in the sense of the parameterizations) and while some vertical transport occurs at this resolution, vertical mixing may be prevented in a larger cell. The consideration of radon and ^{210}Pb vertical cross-sections lead us to

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understand that the differences are not due to few marked individual events, but rather the consequence of the progressive building up of high ^{210}Pb levels over some weeks of hot, unstable and dry weather. We show that, with the parameterizations we use, we obtain a benefit from increasing the resolution, but not to the point to fill the gap with the observed reality; it may be that 0.5° is still to coarse to capture convective updraft, specially near orography, or that our parameterisations fail for these currently exceptional meteorological conditions.

We advocate that this period is thus a very interesting one to critically assess vertical transport in aerosol and climate models, specially as conditions of summer 2003 could become more frequent in the future.

4 Conclusions

In order to study the aerosol distributions and effects at regional scale, it is necessary to use models with sufficiently high horizontal and vertical resolutions. The MOCAGE model has been used in this study to simulate ^{210}Pb as a proxy for submicronic aerosol. We focus on transport and deposition processes only, thus leaving out other source of uncertainty that affect aerosol simulation such as chemical reactivity and emissions.

As a first step, the ability of the model to reproduce ^{210}Pb distributions has been checked at the global scale for years 2001 and 2002, evaluating the simulations against climatological observations from 32 sites all over the world. Then, two simulations have been carried out with the nested version of MOCAGE over Europe, at two different resolutions (0.5° and 1°). Modelled ^{210}Pb concentrations have been compared with the observations from the 5 CARBOSOL sites, located at various elevations (from sea level to 3100 m altitude). As model orography at coarse resolution does not match the real one, model outputs have been considered at the pressure level corresponding to the real altitude of the measurements sites (in standard atmosphere). This method can apply in our case as dry deposition can be neglected. At a weekly time step, results are satisfactory for the period of CARBOSOL, 2002 to 2004. A particular attention was

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paid to the summer 2003, because results were significantly worse (significant underestimation, specially for high-altitude sites) for this season, which was also marked by a very strong heat wave in Europe. Using the same model at different horizontal resolutions, we show some effect (results at higher resolution being better), primarily in link with a better representation of turbulent and convective mixing. As we show that the high levels of ^{210}Pb observed in high altitude are not the result of "fresh" individual convective events (that a model, even with a full-bloom physics and non-hydrostatism, may miss) but rather the consequence of several weeks of accumulation, we think that this is a very interesting case to constrain vertical transport in chemistry and regional climate models, specially as heat wave conditions could become more frequent in the future.

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Table 1. Name and coordinates of observational sites used for the present study.

Number	Name	Latitude	Longitude	Altitude (m)
1	THULE	76.31	−68.46	259
2	BARROW	71.10	−156.30	4
3	KAP TOBIN	70.25	−21.59	22
4	DYE3	65.00	−43.00	2480
5	MOOSONEE	51.16	−80.30	429
6	HELENA	46.36	−112.00	1187
7	REXBURG	43.48	−111.50	1502
8	NEWYORK	40.44	−74.00	56
9	BARBADOS	13.10	−59.50	0
10	BERMUDA	32.40	−64.70	0
11	IZANIA	28.18	−16.30	2400
12	MACE HEAD	53.19	−9.53	0
13	MAUNA LOA	19.28	−155.36	3401
14	MIDWAY	28.20	−177.40	10
15	OAHU	21.50	−157.90	1000
16	ENEWETAK	11.00	162.00	0
17	NORFOLK	−29.20	167.57	10
18	CAPE GRIM	−40.40	144.41	0
19	CHATHAM	−44.0	−176.60	100
20	INVERCARGILL	−46.25	168.20	10
21	GVON NEUMAYER	−70.39	−8.15	10
22	SOUTH POLE	−90.00	0.00	2800
23	MIAMI	25.49	−80.17	4
24	GUAYAQUIL	−2.10	−79.52	7
25	TUTUILA	−14.15	−170.34	77
26	SANTIAGO	−33.28	−70.42	520
27	BEAVERTON	45.32	−122.53	10
28	OKUSHIRI	41.00	139.00	
29	HACHIJOJIMA	33.10	139.80	
30	PERTH	−31.55	115.58	0
31	ORSAY	40.80	2.18	50

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Table 2. Name and altitude (meters) in reality and in the MOCAGE model (1° resolution) for the observational sites of CARBOSOL.

Name	Real Altitude	Altitude in MOCAGE
Aveiro, Portuguese coast	2.5	189
K-puszta, Hungary plain	125	104
Puy de Dôme, Massif Central	1450	844
Schauinsland, Black Forest	1205	533
Sonnblick, Austrian Alps	3106	1692

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Table 3. Average Pb concentrations (in mBq.m^{-3}) simulated by MOCAGE (1°) and measured (in parentheses Hammer et al., 2007) for CARBOSOL observational sites over the whole project.

Name	All	Winter	Spring	Summer	Autumn
Aveiro	0.60 (0.53)	0.80 (0.50)	0.60 (0.48)	0.39 (0.53)	0.60 (0.60)
K-puszta	0.91 (0.80)	1.27 (0.84)	0.96 (0.69)	0.49 (0.79)	1.10 (0.77)
Puy de Dôme	0.43 (0.36)	0.44 (0.18)	0.52 (0.38)	0.36 (0.52)	0.43(0.37)
Schauinsland	0.68 (0.54)	0.84 (0.36)	0.73 (0.51)	0.45 (0.70)	0.73 (0.64)
Sonnblick	0.35 (0.41)	0.31 (0.22)	0.47 (0.39)	0.29 (0.57)	0.33 (0.38)

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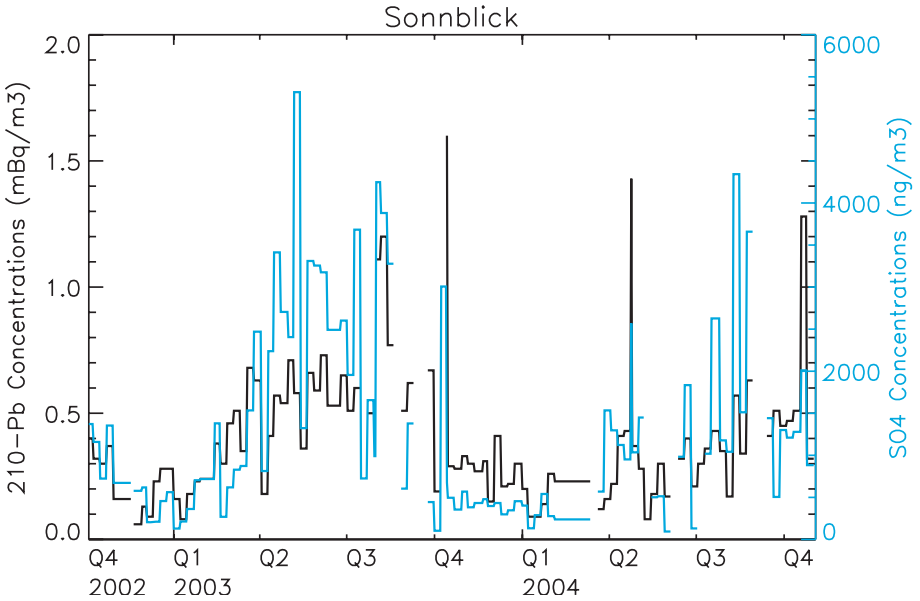

Fig. 1. Observations of ^{210}Pb and SO_4 at Sonnblick during the period October 2002 to September 2004 (CARBOSOL project).

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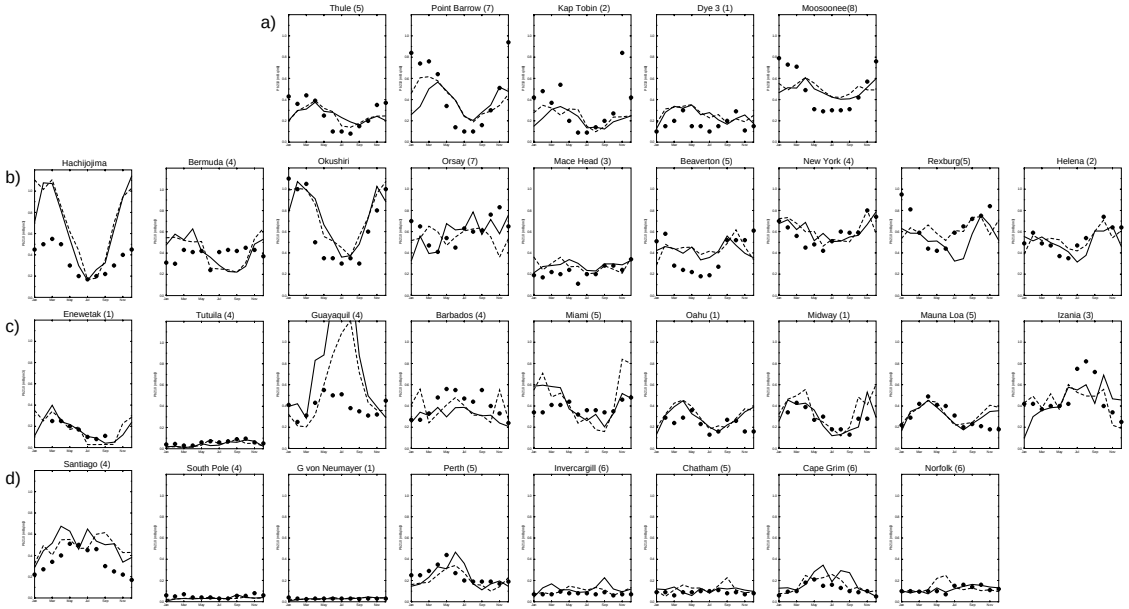

Fig. 2. Monthly mean surface concentrations of ^{210}Pb as simulated by MOCAGE in 2001 (solid line) and 2002 (dashed line), compared to climatological observations (squares), for 31 stations (**a**: Northern Hemisphere high latitudes; **b**: Northern Hemisphere midlatitudes; **c**: Tropics; **d**: Southern Hemisphere mid- and high latitudes).

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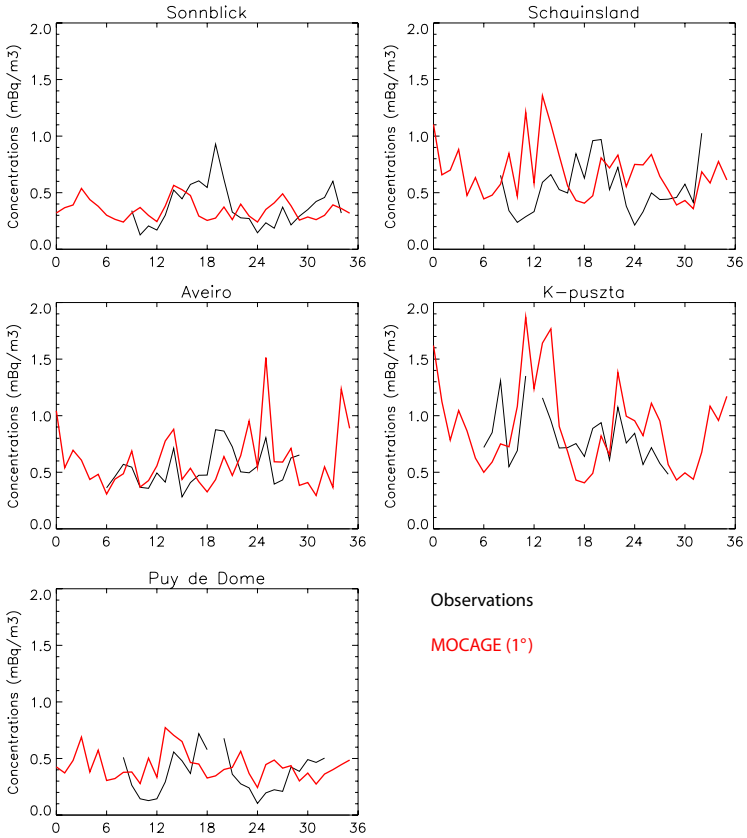

Fig. 3. Monthly surface ^{210}Pb concentrations for the period January 2002 to December 2004 (months 0 to 35): measurements (black), simulations at 1° horizontal resolution (red).

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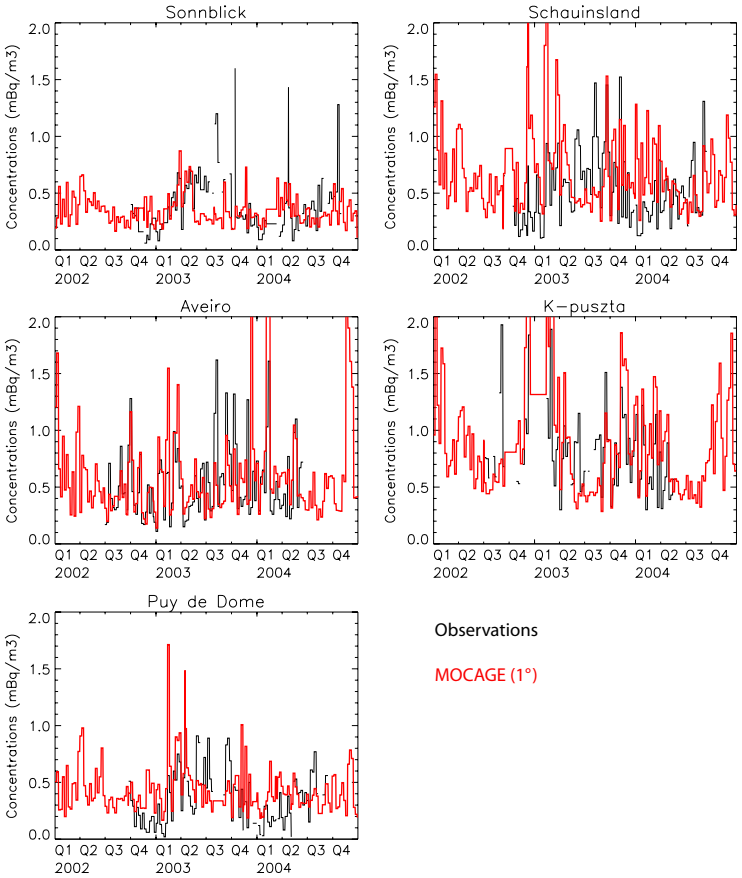

Fig. 4. Same as Fig. 3, but with a weekly time resolution.

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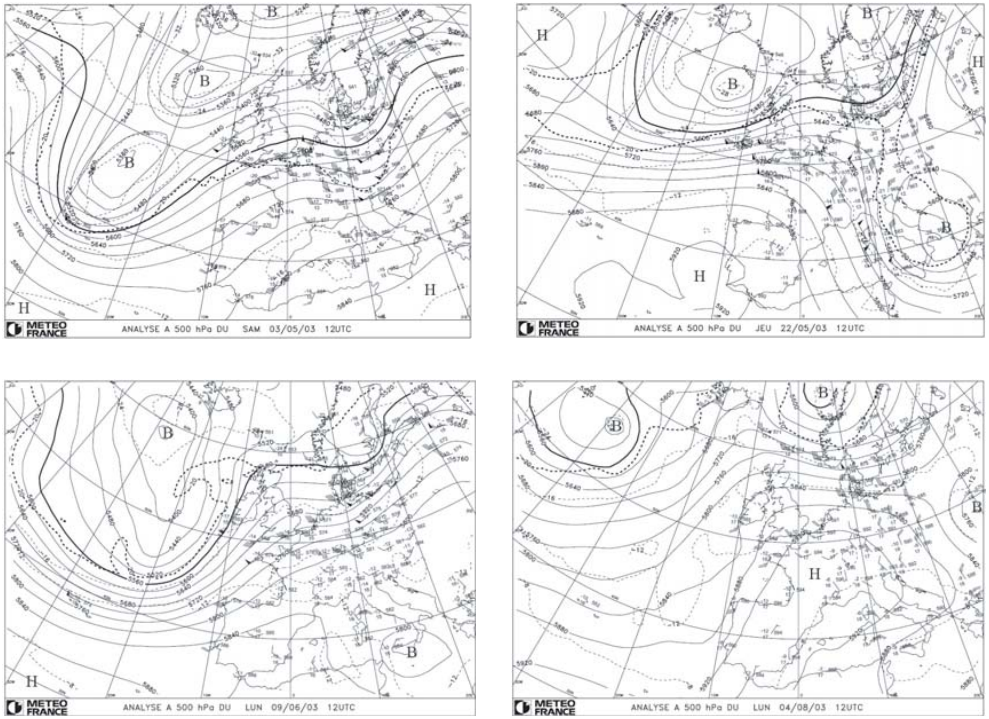

Fig. 5. Geopotential (mgp) at 500 hPa above Europe for four specific days in spring-summer 2003 (12:00 UTC): 03/05/2003 (top left), 22/05/2003 (top right), 09/06/2003 (bottom left) and 04/08/2003 (bottom right).

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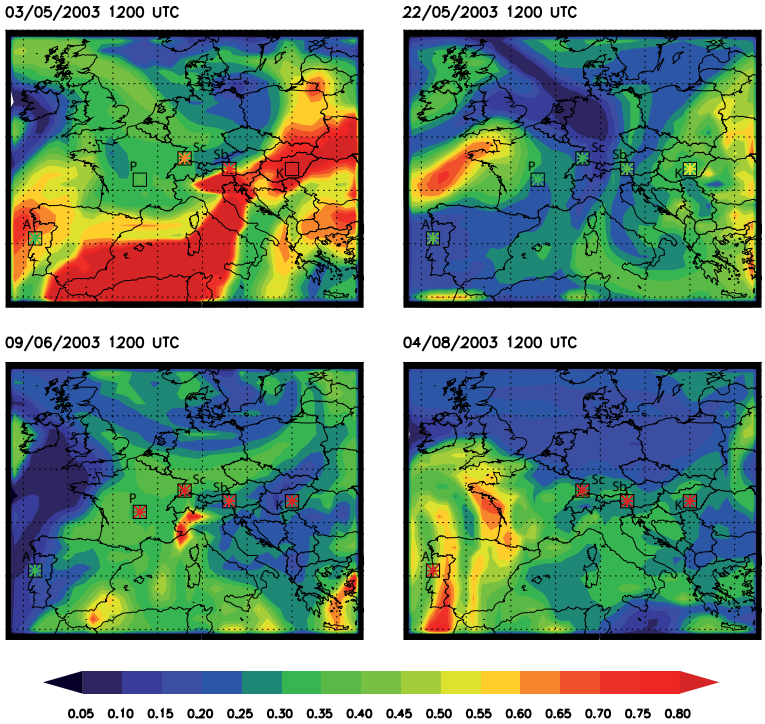

Fig. 6. ^{210}Pb concentrations (mBq.m^{-3}) at 700 hPa above Europe simulated by MOCAGE with 1° horizontal resolution for four specific days in spring-summer 2003 (12:00 UTC): 03/05/2003 (top left), 22/05/2003 (top right), 09/06/2003 (bottom left) and 04/08/2003 (bottom right). The observed values in the 5 CARBOSOL sites are superimposed (star symbols, same color scale as isocontours): A (Aveiro), P (Puy-de-Dôme), Sh (Schauinsland), Sb (Sonnblick) and K (K-Puszta).

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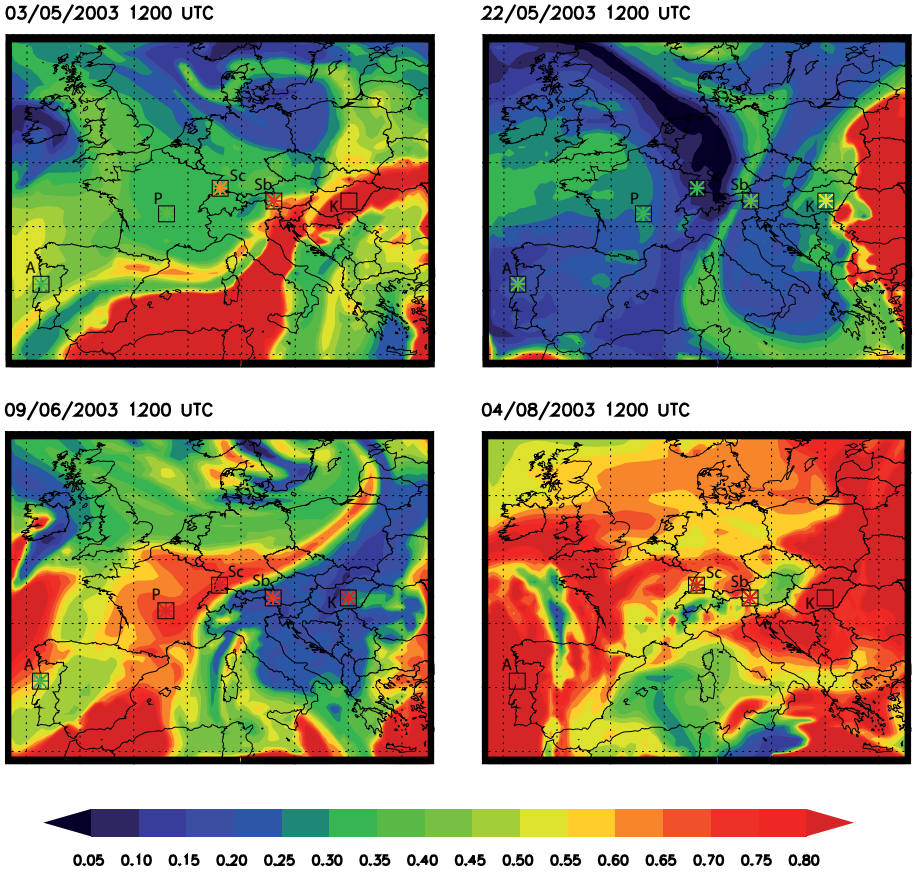

Fig. 7. Same as Fig. 6, but obtained for the 0.5° horizontal resolution.

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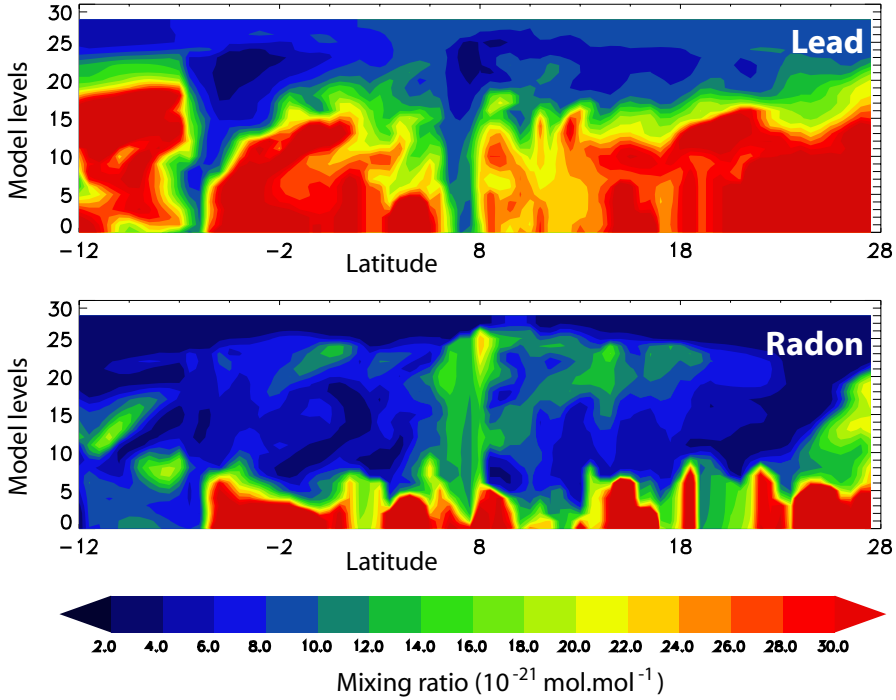

Fig. 8. ^{210}Pb (top) and ^{222}Rn (bottom) mixing ratios: cross-section for 2008/08/04 12:00 UTC at 47° N, corresponding to the latitude of Sonnblick.

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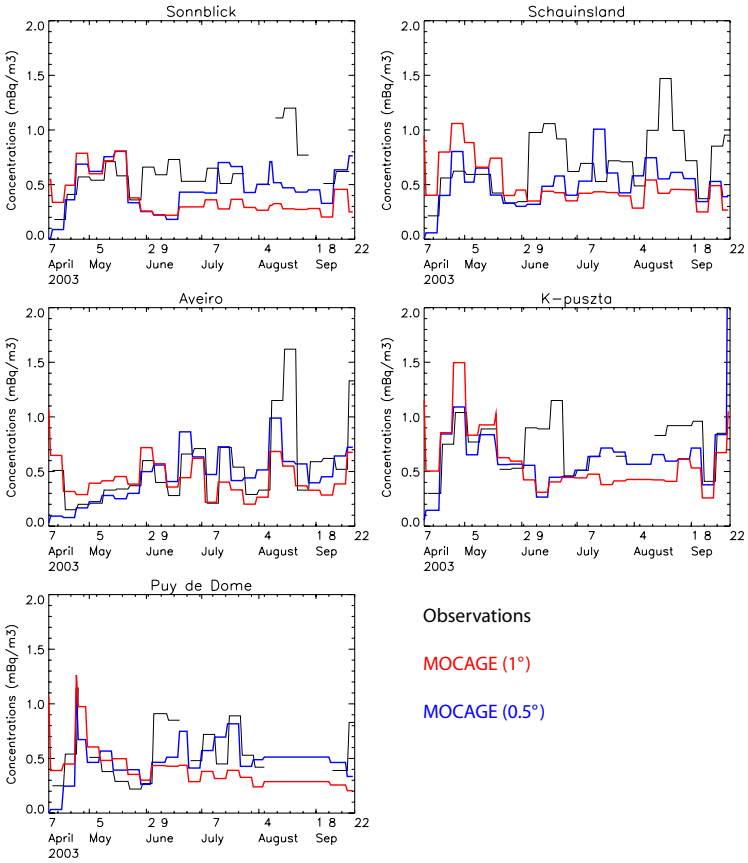

Fig. 9. Weekly surface ^{210}Pb concentrations for the period April to September 2003: measurements (black), simulations with 1° resolution (red) and with 0.5° resolution (blue).

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Evaluation of lead-210 simulations above Europe

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