Inverse modelling of local surface emissions with the chimere-adjoint model: The case of the Paris area during the ESQUIF field experiment

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The aim of this study is to define, test and apply a methodology to optimize surface emissions of anthropogenic species such as NO, at a local scale by using routine measurements of the air quality monitoring networks (measurements available in numerous areas and for long periods). A new methodology in which analyzed maps of concentrations obtained through a kriging technique are used as constraints for the inversion is proposed. The methodology is tested and applied to the Paris area because (i) the kriging technique has been developed for and first applied to this area because of the fully developed AIRPARIF measurement network and (ii) the layout of the area is representative of a simple type of large urbanized area. The NO, emission fluxes were particularly studied because (i) they are ozone precursors with large uncertainties, particularly in their 24-hour time profile (Vautard et al. 2003) and (ii) they are directly linked to NO concentrations that are measured by AIRPARIF and can be analyzed by kriging.

1. Modeling system

The principle of the proposed inversion methodology is displayed on Figure 1. The optimized concentrations provided by the inverse are used to perform a new kriging analysis. The new analyzed concentrations are then used as constraints for a second inversion. This gives the possibility of refining the results of the optimization by iterating several kriging-inversion cycles. The cost function to be minimized is in the form of $J(\epsilon)=(y_{\text{sim}}-y^*)^T R^{-1}(y_{\text{sim}}-y^*)$ where $y^*$ contains the analyzed concentrations, corresponding to each simulated concentration in $y_{\text{sim}}$, and $R$ is taken to be diagonal since the kriging technique does not provide the extra-diagonal terms. Note that information on the first-guess inventory in included in the analyzed concentrations. Therefore, this formulation of $J$ avoids the problem of estimating a "background" matrix.

Our modeling system is based on CHIMERE chemistry-transport model (Vautard et al. 2001) and its adjoint (Menut et al. 2000, Menut 2003). The adjoint approach was chosen because it makes it easy to take the trajectory of the model into account and perform 4D-integration. The kriging technique used here is called "INK" and has been developed by Blond et al. (2003). At any given hour, for any location $s$, the analysis $y^*(s)$ is a correction of the CHIMERE simulation $y^b(s)$ by a linear combination of the innovations $y^*(s_k)-y^b(s_k)$ where $s_k, k=1,...,K$ are the locations of the $K$ measurement values $y^*(s_k)$ provided by the network monitoring stations.

The INK technique and CHIMERE are used by the Paris area air quality network AIRPARIF to produce daily maps of air quality forecasts (www.airparif.org/page.php?article=previś&rubrique=prevision). The minimizer called by the inverse code is N1QN3 (Gilbert and Lemaréchal 1989).

3. The pollution event of the 7th of August 1998

On the second day of the IOP (Intensive Observation Period) 2 of the ESQUIF campaign, a local photo-oxidant pollution event occurred (Menut 2003), in which the local emissions play a
major role (Menut et al. 2000). An ozone plume developed in the South-West of Paris and the difference between upstream and downstream ozone concentrations due to local emissions was at least 50 ppb. The event was simulated with the forecast version of CHIMERE (25x25 cells over 11 vertical levels, covering a 150x150km area). The emission inventory was elaborated by AIRPARIF for the summer of 1998 (described in details in Vautard et al. 2003). It gives the hourly 6x6km fluxes of 16 anthropogenic emitted species among which NOx speculated in 10% of NO2 and 90% of NO.

The studies lead during the ESQUIF campaign have shown that ozone concentrations simulated in the afternoon plume were underestimated by around 8%. In parallel, NO concentrations simulated downtown during the morning were overestimated as compared to measurements, particularly between 6 and 10 a.m. (Menut et al. 2000b).

4. Testing the inversion methodology on academic cases

4.1 Elaborating a realistic academic case

Academic cases based on the 7th of August were run in order to test the inverse code and quantify the limitations of the new methodology. The principle of the inversion of an academic case is summarized on Figure 2.

Series of inversions are performed and an indicator is defined to quantify the quality of the results: the score at one time and location is:

\[ score_x = 100 \times \left( 1 - \frac{x_{opt} - x_{true}}{x_{fg} - x_{true}} \right) \]

with \( x_{opt} \) the optimized emission flux, \( x_{true} \) the true value and \( x_{fg} \) the first- guess value.

4.2 Results

The results for the eight optimized fluxes (four at each of the two hours of the time window) are summarized by computing the average over the eight individual scores, displayed on
5. Application to the real case

5.1 The inverted problem: spatial aggregation

Considering the possible link between ozone and NO miss-estimations, NO\(_x\) emissions from 3 to 10 a.m. are inverted, assuming that (i) the distribution inside the family remains unchanged, (ii) all the other parameters and parameterizations are perfect or at least do not explain the discrepancies between simulation and measurements. To better satisfy the latter assumption, the BLH, that was found to be underestimated by the meteorological data used by the model as compared to measurements made during the field campaign, has been corrected to match the more reliable data.

Since the number of individual fluxes is very high, it is necessary to reduce the size of the problem to match the available quantity of information. First, fluxes less than \(5 \times 10^{11}\) molec.cm\(^{-2}\).s\(^{-1}\) located in the rural areas surrounding the domain are not inverted. Then, zones are defined to aggregate the fluxes to be inverted. In each zone, the same correction is applied to all the aggregated fluxes. In order to satisfy this assumption, the zones are defined by taking into account (i) the intensity of the emission fluxes and (ii) the sensitivity of the concentrations to these emissions. This leads to a dynamical spatial aggregation that makes use of the a priori information available in the emission space. A simple example of aggregation is shown on Figure 5: the four big zones are broken into smaller ones acknowledging for the gradient of intensity in the emission fluxes.
5.2 Results

Spatial distribution of corrections: The inversion time-window ranging from 3 to 10a.m., the spatial distributions of the corrections at 5 and 8a.m. are displayed on Figure 6. This shows that the pattern of corrections is not homogeneous, neither in time nor in space.

At 5a.m., after the optimization, the most intense fluxes located downtown are decreased (multiplied by 0.5 to 0.75) whereas the intense fluxes located outside the city upstream are increased (multiplied by 1.25 to 2.75). On the contrary, at 8a.m., the most intense fluxes are almost unchanged by the optimization with corrections less than 5% whereas less intense fluxes located outside the city are decreased (multiplied by 0.75 to 0.95). Since the situation differs according to the location, the time evolution of the optimized fluxes is studied for two locations representative of the results obtained downtown and in the suburbs.

Optimized emissions downtown: The time evolution of NO emissions in the city are displayed on Figure 7 for the South-West part of Paris that is representative of the whole town. The morning emission peak is reduced from five hours long (from 5 to 9a.m.) to only two hours long (from 8 to 9a.m.).

Since (i) the time profile of the optimized emissions does not show oscillations and (ii) the
A decrease in intensity is consistent with the fact that the first-guess inventory has actually been built for July 1998 (traffic is generally lighter in August in the Paris area), the inversion methodology seems to be reliable for real studies.

Optimized emissions in the suburbs: Four fluxes located in the suburbs upstream Paris were hugely corrected (shown in orange on Figure 6) for one or two hours. The time evolution of NO emissions are displayed on Figure 8 for Tremblay that is representative of these suburban fluxes.

If the peak in the optimized emissions is correctly timed as compared to the measured concentrations, its intensity (more than $4.75 \times 10^{12}$ molec.cm$^{-2}.s^{-1}$) seems unrealistic. It was then assumed that the optimized peak corresponds to a real traffic peak, that has been measured but not simulated and was maybe due to departures on holiday, but the intensity of which is exaggerated probably because of a lack of representativity of the constraints. This problem, that arises for only four isolated fluxes, may indicate that the realism of the corrections decreases with the density of the measurement network.

Impact on ozone concentrations: To validate optimized emissions, their impact on ozone concentrations is examined.

Ozone concentrations simulated in the afternoon plume at 3p.m. were increased by less than 5% by NO$_x$ emissions optimized in the city during the morning (Figure 9). Nevertheless, since first-guess ozone concentrations were underestimated by 8%, the optimization thus reduces the under-estimation to only 3.5%. This shows that the inversion is able to lead to a better simulation of key events.

Conclusion

A new inversion methodology has been designed. It uses an iterative scheme calling on a kriging technique to generate the constraints. Academic cases were elaborated to validate the methodology. Focusing on the Paris area and NO$_x$ emission fluxes, it is shown that the constraints provided by kriging makes it possible to retrieve an emission inventory with a good accuracy, for a wide range of quality of measurements and first-guess inventories. The new methodology is applied to the pollution event that occurred on the 7th of August 1998, second day of the IOP 2 of the ESQUIF field campaign. A dynamic spatial aggregation
The prospects of this work include (i) a quantification of the uncertainty on optimized inventories, possibly by interacting with Monte-Carlo simulations, (ii) the application to other areas and (iii) the use of satellite data instead of/together with surface data.

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References: