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Spatial interpolation of ambient ozone concentrations from sparse monitoring points in Belgium

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ABSTRACT

Due to scientific interest on the one hand and political and regulatory obligations on the other hand the monitoring of ozone in the troposphere is an important issue. To this end, in Belgium as in many other countries, a fixed network of monitoring stations is operated. In order to estimate the ozone concentrations over the whole territory, a model is needed to spatially complement the sparse measurements. This paper describes the development of an interpolation scheme which is aimed at fast operational use. The model uses the population density as auxiliary data to remove a spatial trend due to titration by nitric oxide. The residuals are interpolated by kriging. As a benchmark the inverse distance weighting interpolation method is used with and without the detrending. The proposed model systematically improves the interpolation and makes a significant difference when estimating human exposure to ozone. It is generic in design, easy to implement and flexible to changes in the monitoring network.

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1. Aim of research

Increased ozone concentrations in ambient air have been an issue of research for several decades now, and its impact on human health is still very topical: e.g. Bernstein et al.[1], Maynard[2], Schlink et al[3]. Moreover, due to European regulations (European Community, 2002[4]) the observation and control of ozone in the troposphere also received legal and political importance. To support impact assessments and to meet the requirements of the legislation, the monitoring of ozone is currently very important. Traditionally the observation of tropospheric ozone is performed by automatic analysers and by practical constraints restricted to a number of sampling stations at fixed locations. However, since the real interest of impact assessments is in ozone maps that cover whole regions, a model is needed that spatially complements the observations in the monitoring points.

This paper describes the development of a robust spatial interpolation scheme applied to the territory of Belgium. The main focus is on the adequate incorporation of the titration effect due to NOx pollution, which is very significant in a highly urbanized and industrialized country like Belgium. Since Belgium is not a mountainous terrain, orographic aspect are not taken into account. The goal is to develop a model that can serve in fast operational mode (see section 2.1.), hence we are not considering deterministic atmospheric computer models and we only permit the use of auxiliary data if it is readily available at all times.

2. Description of area and data

2.1. Telemetric network

In Belgium the three regions, represented by the Interregional Cell for the Environment (IRCEL-CELINE)[5], have the task to inform and/or warn the public about the ozone concentrations and to estimate the impact on human health and eco-systems. For this purpose three telemetric air quality networks[†] are operated and ozone concentrations are currently measured continuously (and integrated to 30 minutes averages) at 38 monitoring points (see Fig. 1). At these fixed locations an historical dataset is available, and based on these data a statistical short-term ozone forecaster (for daily ozone maxima) is operational since 1996 [5,6]. However, both the measurements and the forecasts are restricted to the sparse monitoring points and some inductive model is still needed to estimate the full spatial ozone state. Since this interpolation should complement the prediction model, it has to be fast in operational mode.

For this research the data from 1998 to 2003 was used. Some analyses are presented separately for 2003 since it was a year with exceptionally high ozone concentrations. The set of used monitoring stations contains stations located in rural, suburban and urban areas (according to reference [4]). Most of the stations are tagged as background type, some as industrial and traffic; the possible use of this kind of type-classification for interpolation is currently under investigation and will not be the topic of the paper.

[†] The telemetric networks are managed by the Flemish Environment Agency (VMM) in Flanders, the Brussels Institute for Management of the Environment (IBGE-BIM) in Brussels and the Institute for Public Service (ISSEP) and the Ministery of the Environment (DGRNE) in Wallonia.

2.2. Ozone in Belgium

Ozone in the troposphere is a secondary pollutant originating from the interaction of other chemicals and sunlight. The photochemical formation of ozone is influenced by a series of non-linear reactions that were identified in the 1950s by Haagen-Smit[7], among others. The effect of the precursors (being $NO_x \equiv NO\&NO_2$ and volatile organic compounds VOC) on ozone formation depends on their mutual concentrations (Lin et al.[8] , Sillman[8]). Belgium is situated in a region of high NO_x concentrations due to intensive combustion processes. As a consequence the Belgian ozone concentrations in urban areas are usually lower than in their rural counterparts (Dumont et al.[9]); such a "VOC-limited" ozone formation regime is also observed in other north-western and central European urban areas. This knowledge on ozone chemistry is vital to grasp the spatial nature of ozone concentrations in Belgium. On the one hand the ozone phenomenon has a regional scale (of the order of the size of Belgium or larger) related mainly to the meteorological conditions. On the other hand urban NO pollution adds a very local character due to the titration reaction $NO+O_3 \rightarrow NO_2+O_2$ which destroys ozone in urban areas; the resulting NO_2 can reproduce ozone when transported to rural regions. Since this underlying ozone process lacks spatial homogeneity, auxiliary data are needed to assess the spatial structure in the design of an accurate interpolation scheme, e.g. Coyle et al[10], Diem and Comrie[11]. However, with the focus on an operational model, some caution is required. Auxiliary data sets are not always easily available (especially real-time data) and when a multitude of them is used, they can blur the transparency or decrease the stability of the model.

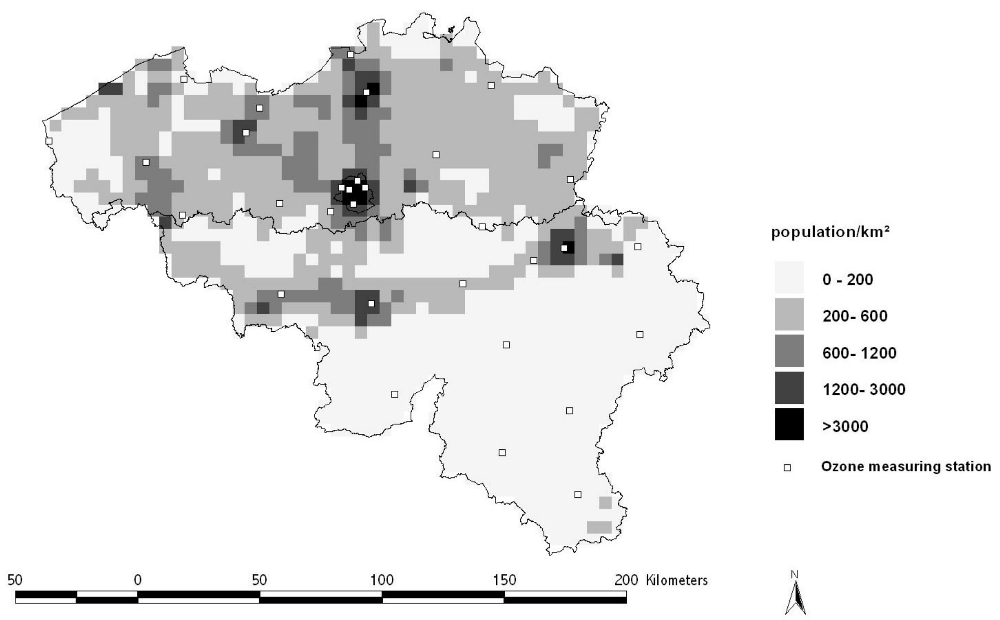


Fig. 1 Population density and location of ozone monitoring points in Belgium

3. Methodology

3.1. The statistical approach

A spatiotemporal process like the formation of ozone, will usually contain spatial and temporal correlations. In this paper however, the temporal aspect of the problem will be neglected. This approximation is justified when the correlations in time are subordinate to correlations in space (which is likely in a dense monitoring network) or to a stationary trend (which will be included). Moreover, in this paper daily maximum values are used and consequently time-shifts are not important if the only effect is a relocation of the maximum within that day. Hence, from now on the notation of time will be omitted and the objective of this research is stated as: estimate the daily maximum ambient ozone concentrations for every cell of a 5km x 5km grid covering Belgium, on the basis of the concentrations measured at the monitoring sites.

A statistical approach to an interpolation problem like this one, usually starts with the definition of a linear estimator: the actual but unknown ozone concentration $O_3(\vec{x})$ will be approximated by an estimator $\tilde{O}_3(\vec{x})$ which is defined as a linear combination of the known measurements at the N monitoring stations located in $\vec{y}_i$:

$$\tilde{O}_3(\vec{x}) = \sum_{i=1}^{N} w_i(\vec{x}) O_3(\vec{y}_i) \tag{1}$$

The interpolation is defined as the optimisation of the weights $w_i(\vec{x})$ for every $\vec{x}$. A conventional way is the method of inverse distance weighting **(IDW)**, see e.g. Isaaks and Srivastava[12], which will be used as a benchmark for our method later in the text. For IDW one first defines the distance between location $\vec{x}$ and $\vec{y}_i$ as $r_i(\vec{x})$ and sets the weights equal to $w_i(\vec{x}) = r_i^{-p}\left(\sum_{j=1}^{N} r_j^{-p}\right)^{-1}$ where p is a positive parameter to be determined by the user. In order to deal with the titration effect in urban areas (described in the previous section) the IDW model was in practical applications for Belgium often accompanied by the following pragmatic approach:

- Remove measurements from urban areas
- Perform an IDW interpolation with the remaining data (power $p = 4$)
- In the urban areas for which measurements are available, locally replace the IDW values by measurements

This algorithm is simple and reasonable, but there are some weaknesses involved in the assumptions on which this technique is based:

1) Ad hoc distance function: In the IDW the weight decay by distance is chosen to be a power law. This is a rather ad hoc assumption, which is not based on the character of the phenomenon. (In the implementation later in the text the value $p = 4$ is chosen.)
2) Clustering: When several monitoring stations are spatially clustered, it is likely that they observe similar concentrations. In a good interpolation model their weight contribution to $\tilde{O}_3(\vec{x})$ should somehow be decreased. In IDW

however the weights only depend on the distance from the measurements, not on the distance between measurements.

3) Spatial homogeneity: Since IDW only includes relative distances, it does not incorporate the possibility of a spatial trend. IDW assumes implicitly that the processes underlying the ozone formation are homogeneous in space.
4) Pragmatic approach: "In the urban areas for which measurements are available, locally replace the IDW values by measurements." This contains two problems. How local should the replacement of IDW values be performed, i.e. what is the influence area of the urban site? And secondly, what to do in urban areas for which no measurements are available?

Problems 1) and 2) will be tackled by replacing the IDW model by a kriging based model. This will be discussed later in section 3.3. An improvement of the errors made in 3) and 4) can be achieved by using the detrending technique described in the following subsection.

3.2. Spatial homogeneity and detrending

Due to differences in the underlying physical and chemical mechanisms, urban areas behave systematically differently from rural. Since most interpolation schemes are based on the hypothesis of spatial homogeneity, this spatial trend poses a problem. Some interpolation techniques can incorporate and estimate a trend, but in this research the chosen procedure is to explicitly determine and remove the trend before the interpolation is performed. In Fig. 2a) a histogram (normalised to a discrete probability distribution) is presented for a typical urban and a typical rural monitoring site in Belgium. The ozone values in this plot and the rest of the paper (unless stated otherwise) are:

$$_{dayM}O_3 = \text{maximum daily 1-hour mean } O_3 \text{ concentration.} \quad (2)$$

Since the summertime is the relevant period for ozone pollution, our study is restricted to the period April – September (1998-2003). The plot clearly shows the negative shift in ozone values from rural to urban sites. To restore the hypothesis of spatial homogeneity, we would like the histograms to be independent of location.

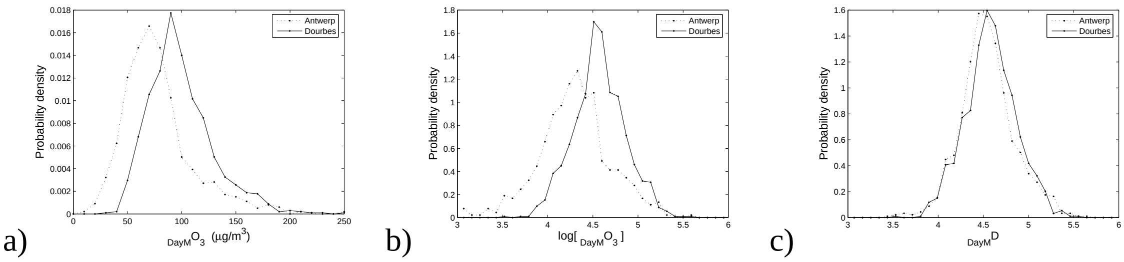
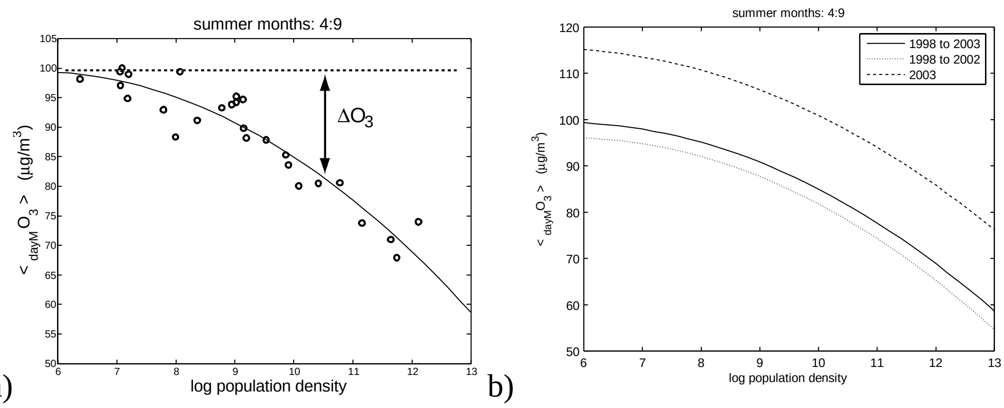


Fig. 2 Probability density (based on data from summers 1998-2003)
a) concentrations before detrending b) logarithmic concentrations before detrending
c) logarithmic concentrations after detrending (Antwerp = urban, Dourbes = rural)

To achieve this goal the spatial nature of NO_x pollution needs to be characterised. It turned out that the population density is very suitable for this purpose. First, contrary to the NO_x data itself, this variable is readily available for a 5km x 5km grid. Secondly, although it is only an indirect characterisation of NO_x the relation with the titration effect is very stringent, as can be seen in Fig. 3 a). In this plot the mean ozone value for week days[‡] of each monitoring station is plotted versus the logarithm of the population density. A clear trend is present and can be fitted, e.g. with a second order polynomial as visualised. Note that this trend is present irrespective of further classification of the stations by proximity of industry or traffic.



a) b)

Fig. 3 a) Measured $\left\langle {}_{dayM}\,O_3 \right\rangle$ for different monitoring sites versus the logartihmic population density together with second order polynomial fit (ozone averaged over week days of summer months of 1998 to 2003).
b) Polynomial fit based on the same data from indicated years.

For every location $\vec{x}$ in Belgium the known population density $\rho(\vec{x})$ gives an estimate of the present ozone shift ΔO_3. This shift is used to detrend the measured or estimated ozone values at that specific location. In this manner, the residual variable at every location has a rural character. To show the robustness of this trend, in Fig. 3 b) the effect of the year 2003 is presented. During that year extremely frequent high ozone concentrations were observed; the effect on the trend is a parallel increase of the curve. I.e. the increase does not depend on population density and consequently the ozone shift ΔO_3 of the trend with or without the data of 2003 is equal.

It is not this residual itself but the logarithm of it that will be used in the interpolation: this detrended variable is denoted as $D(\vec{x})$:

$$O_3(\vec{x}) \quad \rightarrow \quad D(\vec{x}) = \log[O_3(\vec{x}) + \Delta O_3(\rho(\vec{x}))] \tag{3}$$

‡ Fig. 3 presents ozone values for week days during summers 1998-2003. Week and weekend are dealt with separately because NO_x pollution during weekends behaves differently, due to its strong relation with traffic emissions (Vanderstraeten et al.[13]). A similar exercise like that of Figure 2 and 3 was done for weekends, yielding a less curved trend with the same qualitative results: an improved spatial homogeneity.

The log transformation is used because ozone concentrations have a distribution that is similar to a lognormal (Fig. 2a)), hence the logarithmic concentrations have a more Gaussian-like distribution (Fig. 2b)), and Gaussian is best for the interpolation with a linear estimator (1). In Fig. 2c) the result of the detrending is presented. The distributions of the detrended urban and rural site are quite similar; i.e. the probability distributions are less site-dependent and the spatial homogeneity has improved.

3.3. Kriging-based interpolation

Finally the interpolation of the detrended variable (3) has to be performed. A popular and solid technique to this is ordinary kriging, see e.g. Cressie[14] or Isaaks and Srivastava[12]. In this method the weights of the linear estimator (1) are statistically optimised by the use of a spatial correlation that has to be estimated from the data. Usually it is assumed that this correlation between any two $D(\vec{x}_1)$, $D(\vec{x}_2)$ only depends on the relative distance $|\vec{x}_1 - \vec{x}_2|$. The correlation function can to some extend be interpreted as a replacement of the power law of the IDW, with the additional advantage that this correlation is implicitly used to correctly assess the weight of correlated (clustered) monitoring sites. Hence, the kriging method deals with the problems 1) and 2) stated in section 3.1.

In traditional applications of kriging (like e.g. estimations of ore concentrations for mining purposes), one has a multitude of measurements scattered over space without the presence of a time aspect. In such a case, the correlation function is estimated by averaging over many samples in space. In our problem the number of locations is limited to an order of 30, far too low to make a good correlation in this manner. However, since time sequences are at our disposal a different approach is possible. For every pair of monitoring sites a spatial correlation between them can be calculated by averaging over many samples in time. This matter is more profoundly discussed by Szentimrey[15]. The results for every pair of monitoring sites is shown in Fig. 4. A clear decay of the correlation as a function of the relative distance is observable. For the spatial range of Belgium, ± 200 km, a linear approximation seems reasonable and is used.

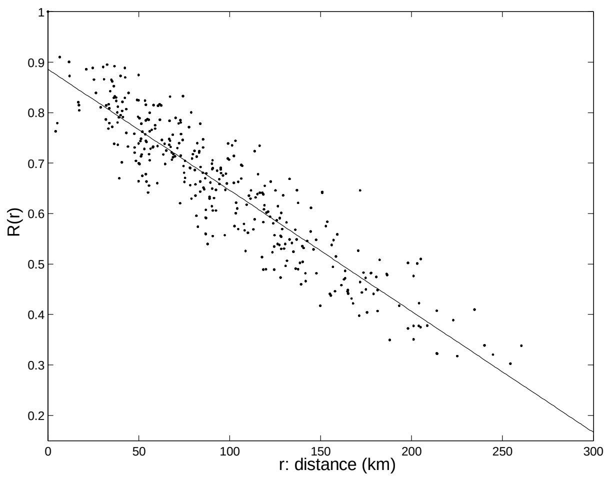


Fig. 4 Correlation between two monitoring sites, as a function of the distance. (The temporal average is based on data from summers 1998-2003.)

4. Results and discussion

4.1. Evaluation of methodology

To evaluate the described methodology, three models are implemented and compared: the simple inverse distance weighting model (**IDW**), a version of this model which interpolates the residuals (3) after the detrending (**R_IDW**) and finally the suggested residual model based on kriging after detrending (**RIO**: residual interpolation optimised for ozone). Obviously, measurements are needed to evaluate or compare these interpolation schemes. Since only point measurements from the monitoring network were available for this task, the comparison is based on "leaving one out": the interpolation methods are fed with all measurements except one, which is estimated by using the interpolation. With this procedure the accuracy of the models can be assessed for each monitoring site.

As a first evaluation criterion the root mean square error between the observed and interpolated values (**RMSE**) is shown in Fig. 5. In this plot only the stations with a history at least starting in 1998 are shown and they are ordered according to increasing population density. This plot shows that generally the error reduces from IDW to R_IDW to RIO. This is most clearly noticeable in densely populated areas. For the residual models R_IDW and RIO the error is less dependent on population density, which is a first indication that the detrending works well.

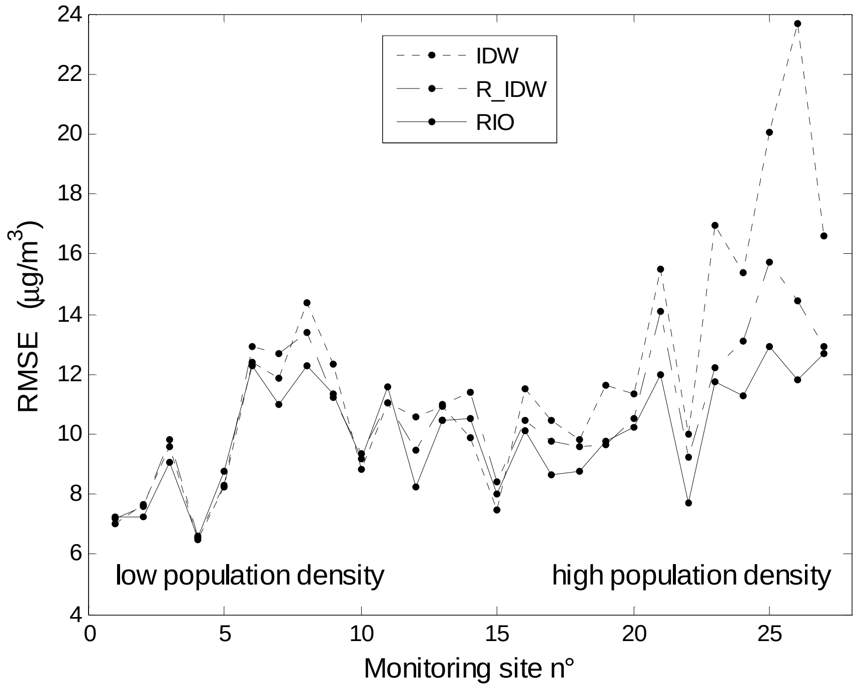


Fig. 5 RMS error of interpolation tools evaluated with "leaving one out". Monitoring sites: ordered according to increasing population density. (The plot is based on data from summers 1998-2003)

For further evaluation the monitoring sites are first divided into a class of high- and a class of low population density. The class boundary is of course rather arbitrary, as a reasonable criterion the presence of a clear titration effect is used. On the basis of Fig. 3 the boundary is set at a logarithmic population density of 10, which corresponds to site n° 20 in Fig. 5. Note that this classification does not coincide with that of the EU directive[4] (urban, suburban, rural) since this is not simply based upon population density. For these two sets of monitoring stations, four set-averaged evaluation criteria are calculated: the RMSE, the mean (signed) error (**ME**), the systematic RMSE (**RMSE_s**) and the unsystematic RMSE (**RMSE_u**). The last two measures are determined from a least square error linear fit of the interpolated values versus the observed. The former is the RMS deviation of the observed values from the corresponding fitted values and gives an indication of the systematic model bias. The latter is the RMS deviation of the interpolated values from the corresponding fitted values, and is consequently a measure for the unsystematic fluctuations of the interpolation model around the linear fit. The result is shown in Table 1.

Table 1 Evaluation criteria of the three interpolation methods for two sets of monitoring stations, averaged for the summers of 1998 to 2003.

	high population density				**low population density**			
	RMSE	RMSE_s	RMSE_u	ME	RMSE	RMSE_s	RMSE_u	ME
IDW	16.9	11.7	11.7	11.2	10.2	4.1	9.1	-0.8
R_IDW	13.1	4.8	12.0	-0.3	10.0	3.8	9.1	-0.5
RIO	11.4	4.5	10.4	-0.1	9.6	3.9	8.5	-0.7

First consider the densely populated areas. From Table 1 it is clear that here the differences between the three models are most pronounced. To begin with, the RMSE_s is much smaller for the two residual models. This shows that the simple IDW was not well-tuned; the detrending described in section 3.2. has seriously reduced the systematic error. Further, on average the ozone shift performed in the detrending was well balanced since the ME is close to zero for both residual models. However, on the basis of RMSE_u there is a conspicuous difference between RIO and R_IDW. While this RMSE_u is similar for IDW and R_IDW it is significantly smaller for RIO, which indicates that the kriging-based interpolation of RIO is intrinsically superior to IDW for the given measurement network. This means that the correlation function that was derived in section 3.3. is adequate and improves the weighting of the different monitoring stations. The fact that both the detrending and the use of kriging improved the model is eminent from the RMSE which decreases from IDW to R_IDW to RIO. All these observations hold for the sparsely populated sites, but are much less distinct. In conclusion, RIO systematically outperforms the two other models, especially in areas of high population density.

To show that the quality of the interpolation models does not decline on years with high ozone concentrations, Table 2 presents the same evaluation criteria determined for the summer of 2003. The values are very comparable to those of Table 1 and the conclusions made above still hold.

Table 2 Evaluation criteria of the three interpolation methods for two sets of monitoring stations, averaged for the summer of 2003.

	high population density				**low population density**			
	RMSE	RMSE_s	RMSE_u	ME	RMSE	RMSE_s	RMSE_u	ME
IDW	16.2	11.2	11.2	9.8	10.5	3.8	9.6	-1.0
R_IDW	14.0	6.3	12.0	-2.0	10.5	4.3	9.4	-0.5
RIO	11.8	5.6	10.0	-1.3	10.2	4.3	9.0	-0.7

4.2. Estimation of human exposure to ozone

To show the significance of the error-reduction presented in Table 1, the methodology is used to estimate the human exposure to ozone. To this end the SOMO35 index[16] is considered. The principle behind this index is that the effects of daily ozone on mortality should be quantified only when the maximum daily 8-hour mean concentration is higher than 35 ppb (70 μg/m^3). Hence, for the quantification of the effect, one considers only days for which the daily ozone concentration (maximum daily 8-hour mean) exceeds 70 μg/m^3, and then only the increment above 70 μg/m^3 is used. This increment accumulated over all days of a year is defined as the SOMO35 index. As the index uses the maximum daily 8-hour mean, the outcome of the accumulation over all days is multiplied by 8 to yield a SOMO35 index expressed in typical (hours μg/m^3) units rather than in (8-hours μg/m^3).

As a first exercise a comparison for the SOMO35 index is made between RIO and IDW for the year 2003 (the last year of the used data set). Both models are applied[§] for every day of the year after which the index is calculated. Subsequently the relative difference (RD) between the two models is determined:

$$RD = \frac{RIO_{SOMO35} - IDW_{SOMO35}}{RIO_{SOMO35}}. \quad (4)$$

In Fig. 6 a visualisation of RD is presented: in the lightest grey areas the RIO estimation of SOMO35 is more than 10% above that of IDW; in the darkest grey areas the opposite holds. When this figure is compared with the population density in Fig. 1, the correspondence is obvious: in densely populated areas RIO estimates a significantly lower SOMO35 index.

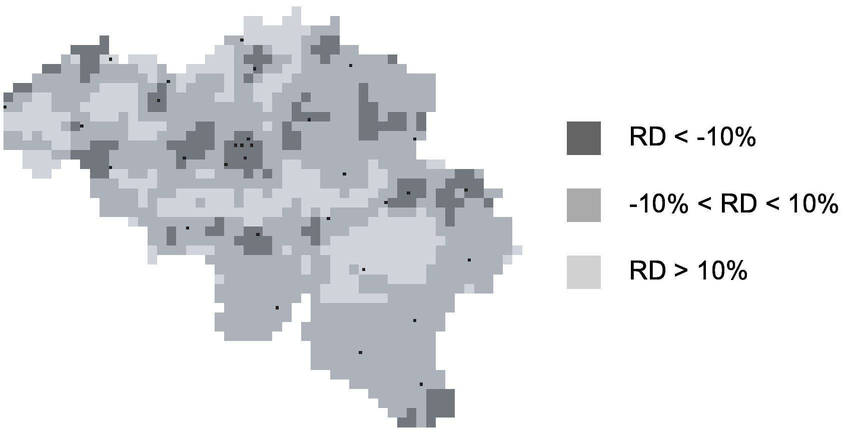


Fig. 6 Spatial representation of the relative difference RD between RIO en IDW interpolation for the SOMO35 index of the year 2003. Ozone measuring stations are indicated by points.

[§] For this calculation a different detrending curve (Fig. 3) is used. Hereto the maximum daily 8-hour running mean is averaged over the summer periods of 1998-2003. Again week days and weekends are separated and both have their own detrending curve.

When spatially averaged over Belgium, the SOMO35 index from RIO and from IDW are equal, hence the difference between the two models is a difference in ozone distribution rather than an overall bias. This difference can become very significant when a weighted average is needed, like for the determination of the human exposure (HE) to ozone. As a measure for HE one can use the local SOMO35 multiplied with the local population density and integrated over the area of interest. The result for the years 1998 to 2003 for Belgium are presented in Table 3. These data clearly show that the RIO estimates for HE are systematically lower than those from IDW. The differences are substantially and are mainly due to lower RIO-values in the densely populated areas. In section 4.1. it was shown that precisely in these areas the accuracy of RIO is distinctly superior to IDW. This leads to the result that for the estimation of human exposure to ozone the proposed methodology is better suited and makes a significant difference.

Table 3 Human exposure (HE) in Belgium estimated by IDW and RIO in units: 10^{12} hours humans.µg/m^3

	HE_{IDW}	HE_{RIO}	$HE_{RIO} - HE_{IDW}$	$\frac{HE_{RIO} - HE_{IDW}}{HE_{RIO}}$
1998	4.73	4.12	-0.61	-15 %
1999	7.13	6.37	-0.77	-12 %
2000	3.77	3.11	-0.66	-21 %
2001	6.17	5.52	-0.66	-12 %
2002	4.90	4.33	-0.58	-13 %
2003	10.48	9.66	-0.82	-8 %

In the assessment of the impact on human health the EU ozone directive[4] uses the WHO guide of 120 µg/m³ as a threshold for the maximum daily 8-hour mean concentration instead of 70 µg/m^3. Using this higher threshold the described analysis of human exposure has been repeated. This results in an even greater relative difference between RIO and IDW, hence the conclusion remains that RIO gives a significantly lower but far more realistic estimation of human exposure to ozone.

4.3. Some remarks

In the current model the population density is used as a measure for titration by nitric oxide. This is of course only an approximation. Firstly, a further distinction between week days and weekends was necessary to quantify the relation with the titration effect. Secondly, the population density is not a perfect measure for all kind of sources of nitric oxide. Probably it is a good measure for domestic heating and also traffic (in Belgium, on a 5km resolution, traffic density is strongly correlated to population density) but e.g. (point) sources of industry are not captured by it. Therefore, the assessment of the titration effect can most likely be improved by using other auxiliary data. Currently, the possibility of land use data (CORINE) is under investigation.

5. Conclusions

The method described above is able to adequately incorporate the NO-titration effect in a spatial interpolation model for ground level ozone concentrations, provided that weekends and week days are dealt with separately. Population density is used as a best approximation for the spatial distribution of the NO-titration. The method is designed for use in fast operational mode. Therefore the model is kept robust and easy to implement. It is very flexible to the addition or removal of monitoring stations, since no extra tuning or training is required. The auxiliary data (population density) is easy to acquire and does not require frequent updating. Despite its simplicity it significantly improves ozone assessments, especially for human exposure, in comparison with a standard IDW model. RIO is currently producing on-line hourly ozone maps for Belgium[5] and is immediately applicable in other VOC-limited and non-mountainous areas.

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