Atmos. Chem. Phys. Discuss., 10, C5890–C5897, 2010
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multi-annual changes of NO_x emissions in megacity regions: nonlinear trend analysis of satellite measurement based estimates" *by* I. B. Konovalov et al.

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Received and published: 30 July 2010

Response to the comments of the anonymous referee #2

We are thankful to the reviewer for the generally positive evaluation of our paper and helpful comments. Our point-to-point responses are given below.

Comment: Inverse modeling. An assumption in Section 3.1 is that there is a linear relationship between the NOx emissions and the NO2 column density. This is demonstrated in Figure 3 quite nicely. However, London seems to have rather low noise and pretty consistent downward trend. It would be valuable to show that this is true for all of the mega-cities investigated here.

The same analysis was repeated for all of the cities, but no noticeable nonlinearities were found. To keep the paper length within reasonable limits, results for only two more cities (Moscow and Istanbul) representing different geographical zones are shown in Fig. 3.

Comment: Method description... My specific questions are 1. How is the neural network configured?

This is a somewhat puzzling question because the configuration of the network was presented explicitly by Eq. (5). To simplify the understanding, the statement "we use a neural network (of the perceptron type)" in line 20 of the reviewed paper is changed for "we use a three-layer feed-forward network with a sigmoid transfer function".

2. A standard perceptron network gives binary values, but the approach described here yields real value – explain?

It is true that a classical perceptron invented in 1957 gives binary values. However, nowadays the network employed in this study is commonly called "multilayer perceptron" (see the references given in the first paragraph on page 10940), although it yields real values. To avoid a possible confusion, the term "perceptron" is omitted in the revised paper.

3. What happens to beta_0 and beta _1 in Equation 5? These terms disappear from the right-most expression.

These terms describe a linear part of the trend, while the right-most expression defines the nonlinear transfer function (g) of a neural network. Accordingly, these terms should not appear in the definition of the nonlinear part of the trend.

4. The mega-cities are divided into those where a linear trend can be estimated and those where only a non-linear trend can be estimated. How is this determined? And how is the green line marked "interannual change" estimated? How is this different than the purple line marked "retrieved trend"?

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The trend is linear in two cases: (1) the algorithm returns w_max=0 with all considered numbers of neurons (N=1 and N=2, see a discussion concerning the choice of the maximum number of neurons in the last paragraph on page 10941). This means that all weight coefficients for non-linear trend components are zero in accordance to Eq. 7 on page 10940. Or (2) the optimal number of neurons defined in accordance to the procedure explained in p. 10941 equals zero, N=0. This point is clarified in the revised manuscript.

The points on the green line (which represent the rate of interannual change of NOx emissions) are obtained as the difference between values of the retrieved emission rates they are shown by the blue line) in two neighboring years. We took into account that the obtained NOx emission estimates are more accurate in points supplied with input data, rather than between them. The points (on the green line) are linearly connected for the better visibility. An equation showing our definition of the rate of interannual change and a corresponding remark are added in the revised manuscript.

Comment: Noise estimates. Equation 10 describes the method for estimating the noise. To what extent is this tied to known uncertainties in the satellite retrieval or errors in the inverse modeling? I appreciate that this question is beyond the scope of the paper, but is there anything that can be interpreted from the site-to-site differences in the estimated noise? It seems like this in an area where this method could also make an important contribution.

Strictly speaking, the noise estimated within our algorithm does not directly relate to the noise in satellite measurements because we analyze the time series of NOx emissions estimates which were obtained by combining satellite data with simulations as specified by Eq. (3). Accordingly, the noise level depends not only on uncertainties in satellite data but also on model errors and on contribution of the “background” NO2 column amounts. Moreover, the noise estimated in this study includes only the random (varying) part of model and measurement errors, because a systematic error (a bias) which is much more difficult or even impossible to evaluate does not influence the trend

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estimate. Therefore, the interpretation of site-to-site differences in the noise level is not at all obvious. Nonetheless, to provide the reader with more information, values of the standard deviation estimated in accordance to Eq. 10 (they are typically about 0.1) are given in Fig. 7 and 8 of the revised manuscript. An additional comment clarifying the meaning of the estimated standard deviation is added after Eq. 10.

Comment: Also, the uncertainty bounds on the retrieved trends in Figure 7 seem quite small.

Unfortunately, the reviewer has not explained why the uncertainty bounds seem to be quite (too?) small. In fact, the estimates of uncertainties in linear trends shown in Fig. 7 in terms of 68.3 percentile were verified using a standard option of the popular commercial graphical software (Grapher 7.0) and also by means of the analytical estimation (which can be done only in the case of linear trends). Also, our estimates of uncertainties in linear trends of NOx emissions are comparable to uncertainties in trend in NO2 columns reported in the paper by van der A et al., 2008. In particular, as it was noted in the revised manuscript, the uncertainty of the trend reported by van der A et al. for Teheran is only 1 percent per year, while in our study we got 1.5 percent per year.

Comment: Can you show that if the method is repeated using the surface observations, the trend falls within the bounds shown in Figure 7?

As far as we know, the trend estimated with two different sets of independent data should not necessarily fall within the uncertainty bounds estimated for each dataset. The reason is that the uncertainty bounds are always based on probabilistic estimates. That is, even if the uncertainty bounds represent 99th percentile of the assumed probability distribution of the considered estimates, it is still possible that the real value is actually outside of these bounds. Nonetheless, after applying our algorithm directly to surface NOx observations in Paris, we got a similar accelerating negative nonlinear trend. Specifically, the rate of the surface NOx concentration change between 2000

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and 2001 is -2.90 percent per year, which is inside of the confidence interval of the rate of the NOx emission change in the same period ([-3.5;1.3] percent per year), as estimated with satellite data. Similarly, for the period between 2007 and 2008 we got the interval [-10.1; -4.3] percent per year with satellite data and -5.42 percent per year with surface data. Additionally, using NOx measurements in London, we got the trend to be -4.2 percent per year, which is quite consistent with our satellite measurement based estimate of this trend (-4.47 $\pm$1.07). The corresponding comment is added in section 4.2. A similar comparison cannot be easily done in the case of other cities (specifically, Madrid and Milan) because the available time series of surface measurements are shorter than time series of satellite data. Nonetheless, the piecewise linear regression analysis presented in Fig. 9 of the reviewed manuscript confirms that the trends in surface observations are consistent with the estimated NOx emission trends within estimated uncertainty limits.

Comment: Is there a different way to falsify or evaluate the quality of the retrieval uncertainty?

To the best of our knowledge, the only objective way to estimate the uncertainty intervals in situations when analytical solution is impossible is based on the Monte-Carlo experiments. That is, this is the same way which was used in this study. As it was noted above, we made sure that analytical estimates of uncertainties and our Monte-Carlo experiments yield practically identical results in a linear case.

Comment: It is preferable to refer to the original definition of megacities, rather than wikipedia I think you can simply say “as defined by the United Nations.

The reference to Wikipedia is removed from the revised paper.

Comment: What is the meaning of “preliminary convoluted” here? Is it preliminary in that a more complete analysis is performed in a separate part of the method? If this is a technical term, please provide a reference that describes the method.

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No, “preliminary” is used to describe the order of operations. The convolution is described by Eq. (1). To avoid misunderstanding, the word “preliminary” is removed from the revised paper.

Comment: 10931:12. This sentence is unclear: “Such an evaluation of sc implies that the change of NOx emissions between the years 2002 and 2003 may be disregarded in comparison with the maximum change of emissions during the whole period of 13 years.”

This sentence is improved in the revised manuscript.

Comment : What are the units of sc? Grid cells?

Yes

Comment: Later in line 20: “about 95 percent of the signal” how does this follow from the calculation described above?

This follows directly from the well–known properties of the Gaussian distribution. We mention this analogy in the revised manuscript.

Comment: Please describe method of NOx measurement. The most common technique is chemiluminescence, which has been shown to have biases in urban environments (Dunlea et al., 2007). If applicable, how do you interpret your results in light of these errors?

We have discussed already this problem in our earlier paper (Konovalov et al., 2008). Here is the citation from this paper:

“Note that chemiluminescence analyzers which are employed in the UK automatic network may overestimate the actual concentration of NO2 because of interference of non-NOx reactive nitrogen (NOz) species. For example, Dunlea et al. (2007) found that, during the MCMA-2003 field campaign in Mexico City (April 2003), the interference of NOz species resulted in average NO2 concentrations measured by the chemilumi-

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nescence monitors up to 22 percent greater than that from co-located spectroscopic measurements. Steinbacher et al. (2007) found that, on the average, only 70–83 percent of NO2 measured at a non-elevated rural site could be attributed to real NO2. We have no specific information about possible artifacts in the considered NOx measurement in Great Britain, but it seems probable that they should be much smaller than those mentioned above. Indeed, the majority of the selected NOx monitors (19 out of 21) are located in urban areas, and, therefore, the ratio of concentrations of secondary pollutants containing nitrogen (such as HNO3 and alkyl nitrates) to those of primary NOx pollutants at the selected AURN sites should be significantly smaller than at the rural sites considered by Steinbacher et al. Similarly, the climate in Mexico City is warmer and actinic fluxes larger than in Great Britain, and thus it is more favorable to oxidation processes. A bias which these artifacts may cause in the measured negative trend in NOx concentrations in UK is likely positive, because concentrations of OH and O3, which are responsible for the oxidation of NOx species, are likely to increase in urban sites as NOx emissions decrease. The magnitude of this bias is quite difficult to assess; most probably, it is less than 10 percent of the measured trend. Such bias cannot change any conclusions of this study."

Most of this discussion is applicable directly to this study. Accordingly, the discussion of this problem in the revised manuscript is very brief.

Comment: Table 1 Please correct inconsistent capitalization in "Monitor's Type" column.

It is done

Comment: anyway Please consider removing the use of "anyway" and "besides" as a transition between thoughts. It is distracting, because in conversation, "anyway" is often used to transition to a different line of reasoning. In most places they can be removed without any loss of meaning.

We follow this recommendation in the revised manuscript.

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Comment: Equation 7. I am having a very difficult time parsing the vertical bars in this expression. Are the vertical bars around w for absolute value? What are the other vertical bars for? What does “const” mean in this context? I’m confused because while wmax is a constant, pa(w) is not, correct?

We are sorry for these difficulties, but we used only standard mathematical notations. Specifically, vertical bars around wmax denote absolute value and other vertical bars denote condition (if). In the revised manuscript, we use the word “if” instead of the corresponding vertical bars. As usual, “const” means a arbitrary positive constant (whose value is not important in a given context); pa(w) can either equal const or be zero, depending on w and w_max. We do not see here any contradiction. In the revised manuscript, an additional explanation is added after Eq. (7).

Comment: 10942:19 “It does in fact not influence their calculation” whose calculation? Awkward sentence, please re-phrase.

The sentence is re-phrased in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 10925, 2010.