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Review Article

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Environmental-Persistent Free Radicals Prediction by Neural Networks

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Abstract

Air pollution is recognized as a critical threat to human health. The link between the inhalation of fine particulate matter (PM) in combination with toxic gases and the resulting health effects is well documented. The health damages were proven to be interrelated with the free radicals caused by air pollution. Free radicals cause a destructive chain reaction by taking an electron from a stable molecule and converting it into an unstable one. Environmentally persistent free radicals (EPFRs) were not considered until the last ten years as an important part of the air pollution composition. EPFRs are a special class of free radicals with a long half-life, contributing to the development of aging-related diseases in living organisms, as well as harmful oxidative effects in lung tissue.

The work is grounded on database of the routinely measured air quality parameters (air pollutants O3, CO, NO2, SO2, PM2.5) and corresponding EPFR data to develop a model for EPFR prediction. An intelligent instrument is used to perceive the pollution information and, on this basis, EPFR values without direct measurement were calculated. Applying the above approach, an average deviation of 0.73 was accomplished.

Keywords: Air Pollution, Artificial Neural Networks, Environmentally Persistent Free Radicals

1. Introduction

The concept of persistent free radicals was first introduced in the 1950s [1]. In 2007 a precise mechanism of EPFR formation was proposed by Dellinger et al. [2]. The core idea in describing these mechanisms is the key role of the substituted aromatic compounds acting as molecular precursors through the reaction with transition metal oxides. Chart with the steps of reaction of different precursors with transition metal oxides in forming diverse EPFRs is given in Fig.1. It is believed that the transition metal oxides are acting as catalysts for the reactions of different organic compounds and this way enhance formation of EPFRs on the corresponding fine particles. Among the heavy metal oxides with catalytic abilities to form EPFR it has been shown that ZnO has higher potential [3]. There are evidences [4, 5]. That some metal oxides are forming stable metal-EPFR complexes on the fine particle surfaces, which do not react rapidly with air and can spread the negative effects over long distances. In this sense, a common form of air pollutant, namely the PM could have significant toxic effects on human health [6].

There are several studies confirming that EPFRs interact with oxygen molecules and producing reactive oxygen species, which boost the negative EPFRs human health effects [7- 9]. The above mentioned example of EPFRs – O2 interaction shows that knowledge on the air constituents is essential to evaluate the health risks of EPFRs in atmospheric particulate matter [10].

In assessing the adverse effect of EPFRs it is of great importance to notice their lifetime in the environment as it could directly influence the extent cells damaging [11]. The typical lifetime of short-lived free radicals (such as hydroxyl and superoxide radicals) is approximately 10-9s, while EPFRs can exist for months or even years [12, 13].

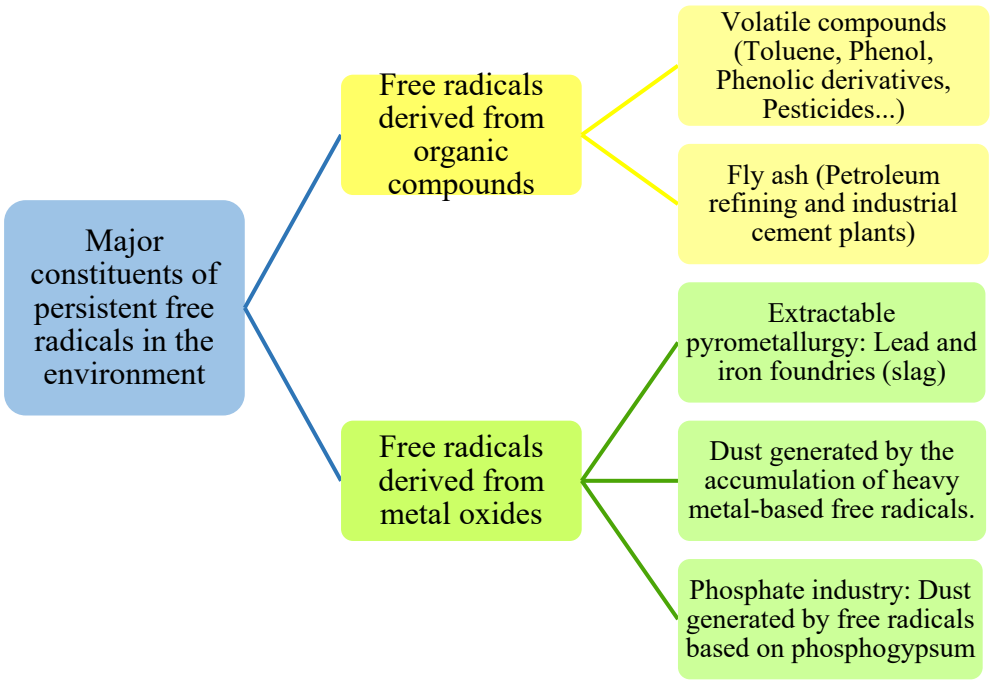

Figure 1: Principal Sources of Environmental-Persistent Free Radicals.

The toxicity of EPFRs is interrelated to their ability to produce •OH radicals, which further increases the formation of other reactive oxygen species (ROS), such as peroxyl (RO2•) and alkoxyl (RO•) [14]. These ROS can induce oxidative stress in biological systems [15, 16]. EPFRs has been detected at high concentrations in atmospheric particulate matter (PM), iron ore, clay, micro plastics, and combustion by-products derived from wood, raw coal, bio-char [17] and products of the metabolic processes of organisms as well sewage sludge [18, 19]. It was shown that in road dust (fine particles and depositions), the polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) are EPFR precursors [20- 22] (Fig. 2).

The most common method to measure EPFRs is electron paramagnetic resonance spectroscopy (EPR) or electron spin resonance (ESR). EPR is well accepted technique used to study objects containing radicals. The EPR analysis are performed through two approaches, namely, by the direct determination method and the solvent extraction method. Both are applied to studies on the EPFRs measurements in the atmosphere [23]. However, this method has some drawbacks, mainly related to quite sophisticated apparatus used and need of well-trained labor.

Several studies researched and reported a relation between commonly measured air pollutants and EPFR. Significant correlations were found between EPFRs and SO2, NO2, and the thermally derived OC3 and OC4 carbonaceous components. Such pollutants have been found in coal-fired and traffic products, representing important sources of EPFRs in PM2.5. In addition, there are proofs that EPFRs are positively seasonally correlated with O3.

Khachatryan et al. (2014) have shown that resonance-stabilized, environmentally persistent free radicals (EPFRs) (semiquinone, phenoxyl, cyclopentadienyl, etc.) can be formed on the surfaces of fine particles. The authors hypothesized that the redox cycling of adsorbed EPFRs is a source of reactive oxygen species (ROS), such as hydroxyl radicals (•OH), superoxide anion radicals (O2•-), and hydrogen peroxide (H2O2) [24].

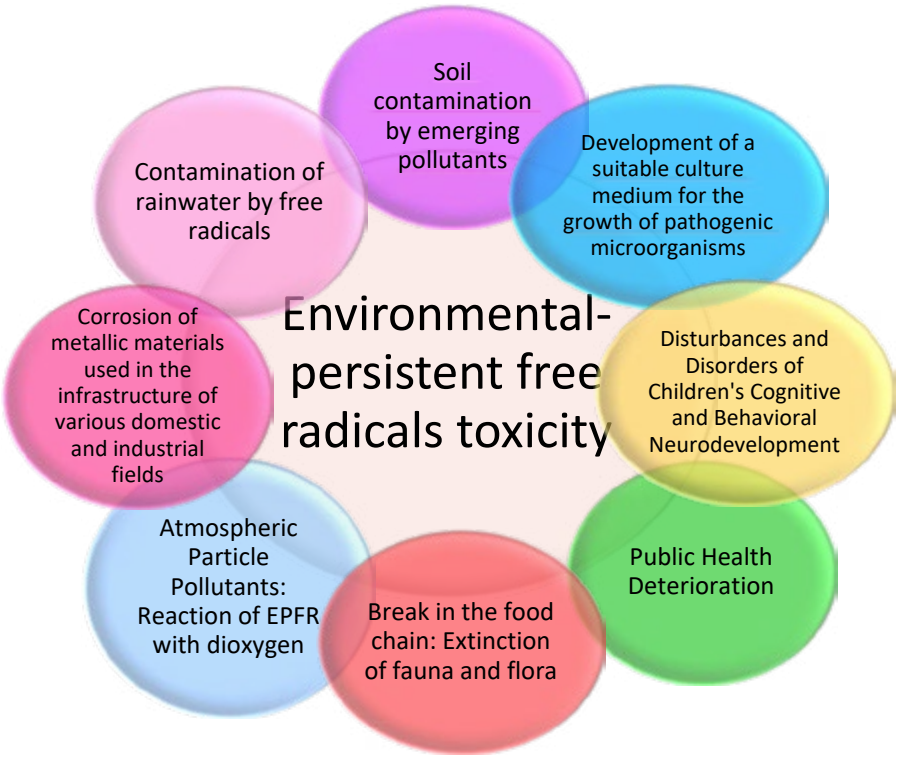

Figure 2: Impacts of Free Radicals on the Environment and Human Health.

Secrest et al. revealed that several chemical species in PM2.5 are intrinsically correlated [25]. They identified groups of linear correlated chemicals as the main contributors to PM2.5 exposures and used the data for exploratory factor analysis.

The influence of atmospheric parameters such as temperature, ozone concentrations, nitrogen oxides, and particle acidity, in combination with other side parameters such as precipitation, wind speed, and humidity on the behavior of airborne EPFR was also recently studied [26]. Xu et al. applied a multiple linear regression aiming to assess the influence of the above parameters on EPFR concentrations. The model predicts well the EPFR level during the heating/non heating periods. The data analysis performed had shown that the levels of EPFRs correlate positively with levels of, carbon monoxide and nitrogen dioxide. [27].

In order to prove the correlation between PM concentration and the atmospheric EPFR levels, a three-dimensional air quality model based on interrelation between PM2.5 and ozone was created [28]. The linear dependence on ozone is proved both at heating periods and at lower temperature periods [29].

Many of the articles comment on the issue of choice of method of analysis. Most of them recommend the use of ANN over regression analysis. This is clearly shown in an analysis in, which discusses key issues and challenges in the application of these techniques in water, including the ethics of these technologies for decision making in water resource management and governance [30]. Finally, recommendations and future directions are given for the application of the different deep learning methods and models in the fields of hydrology, water resources and fine particulate pollution.

Realizing that the free radicals level is influenced by several components of the atmosphere in a relatively complicated relation demonstrates the need of a new approach integrating and assessing the impact of the main air pollutants on free radicals' behavior. Machine learning through artificial neural networks (ANN) has become a crucial tool that can predict the chemical behavior of various chemical substances. The mathematical apparatus of neural networks has been successfully used in various information based industries. In our case, ANN has been applied to model tropospheric ozone in order to predict its concentration [31]. In this study the values of temperature, humidity, and wind direction are used as factors influencing the behavior of some substances. The accuracy of the prediction of ANN against the measured data has been evaluated using the so called Index of Agreement (IOA). Another Artificial Neural Network application was developed to predict the Smoke Point (SP) of different fuels and their blends [32]. A positive linear relation was observed between the measured and predicted SPs.

This paper aims to present some findings on this problem obtained by application of an Artificial Neural Network as an approach to study the interrelation of commonly measured in air pollutants such as O3, CO, NO2, SO2, and PM2.5) and the EPFRs in the

atmospheric air. Data for the study were taken from literature sources. They are presented in Appendix 1 (at the end of the text) and consist of 207 measurements.

2. Materials and Methods

2. 1. Neural Networks

One of the major parts of artificial intelligence is borrowed from basic biological structures. Neural networks are an instrument that borrows their structure from the structure of the neural networks of the human brain, Fig.3.

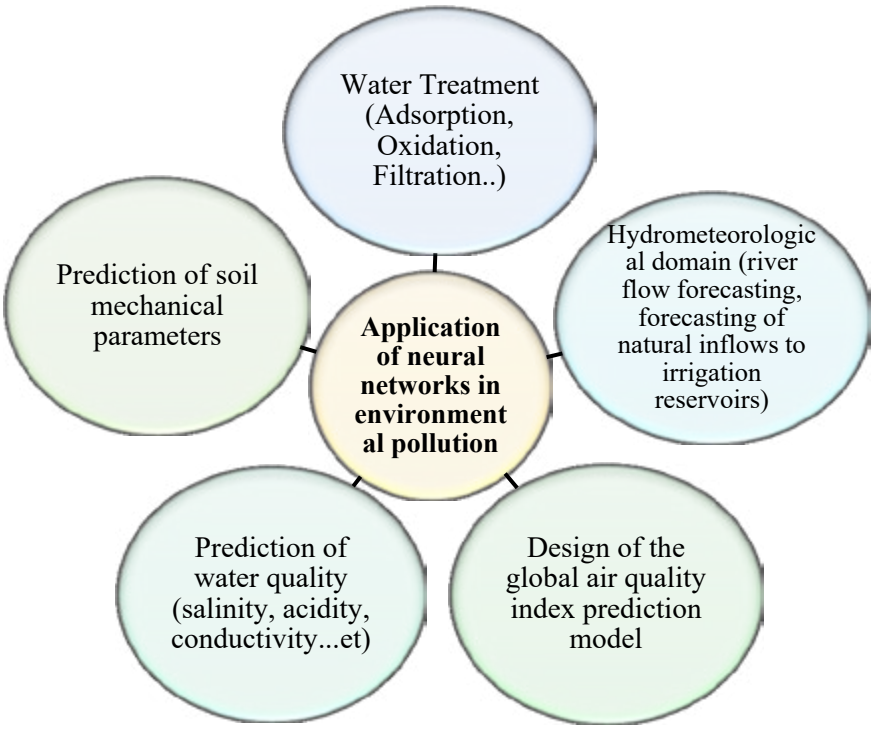

Figure 3: Environmental Application of Neural Networks.

The basic theory and algorithms of deep learning provides the connections between neural networks and traditional machine learning algorithms [33, 34]. The concept of index matrix, the related extended matrix calculus, intuitionistic fuzzy sets based on experimental data are the core of several types of neural networks recently developed [35].

It is well known that artificial neural networks can perceive information "that is not visible" and transform it into their weighting coefficients. The figure shows the abbreviated recording of a two-layer neural network (Fig. 4).

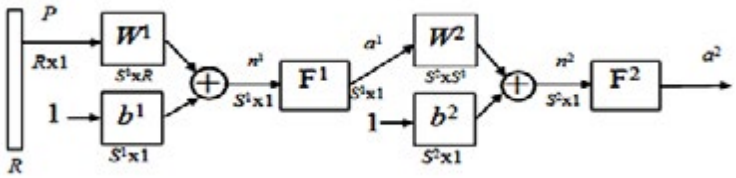

Figure 4: Abbreviated Notation of a Two-Layer Multi-Layer Perceptron

In the two-layered neural networks (4), one layer's exits become entries for the next one.
Eq. (1) describes this operation as follows:

$$a^2 = f^2\left(w^{2f^1}(w^1 p + b^1) + b^2\right) \qquad Eq.(1)$$

where:
- a^m is the output of the mth layer of the neural network for m = 1, 2;
- b is the input bias of the neuron;
- w^m is a matrix of the weight coefficients of each of the mth layer entries;
- f^m is the transfer function of the m^{th} layer.

3. Results and Discussion

The proposed neural network has a structure of five inputs (Fig.5 and Fig.6), eighteen neurons in the hidden layer, and one output (3). Five parameters are submitted at the inputs of the MLP: O_3 (mg/m_3), CO (mg/m^3), NO_2(µg/m^3) SO_2(µg/m^3), PM2.5 (µg/m^3), and at the output of the neural network, the concentration of EPFRs.

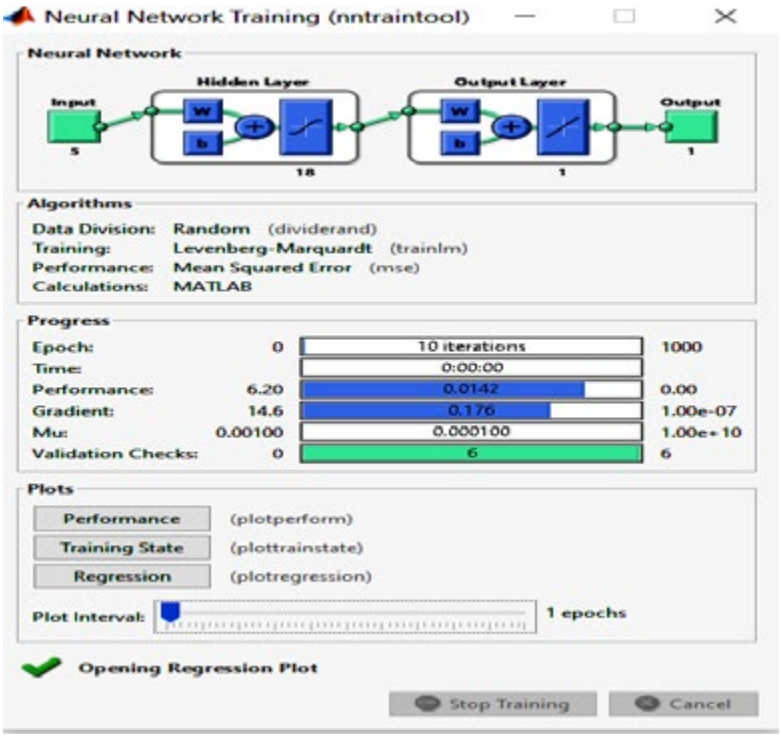

Figure 5: Structure of the Neural Network

The neural network was trained with data from 51 parallel measurements of all data conducted in the period March-December 2018 with the following parameters: structure of the MLP 5:18:1; training epoch limits 1000. The data are divided into three parts: vectors for verification,vectors for testing and vectors for training. It should be said that the verification vectors are not involved in the training process, but are used to simulate real data.

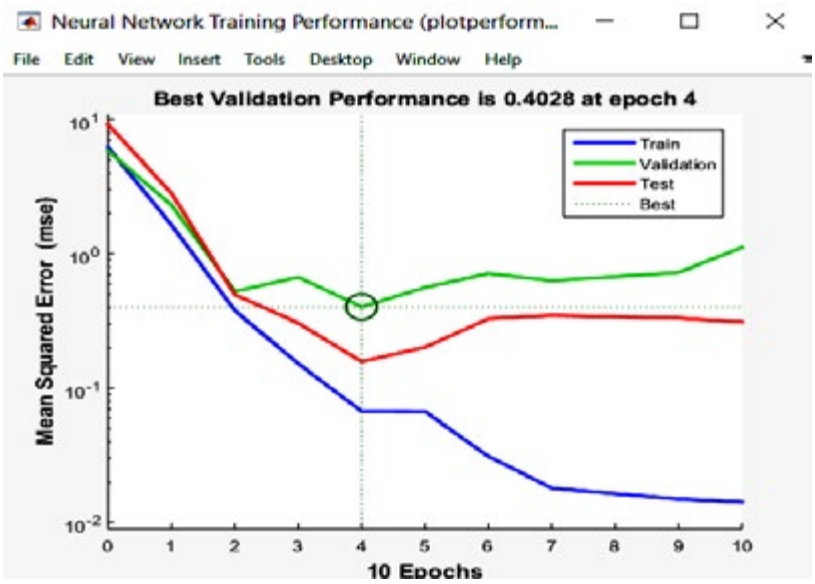

Figure 6: Learning Process of MLP

The achieved parameters are the following: 10 iterations, performance 0.0142 (mean square error), minimum gradient 0.176, the best validation performance is 0.4028 at epoch 4, and the best performance is 0.0142 at epoch 10. The coefficients of regression are: for the training R=0.89959, for the validation R=0.36578, for the test R=0.46201, for all processes R=0.73091 (shown in Fig.7).

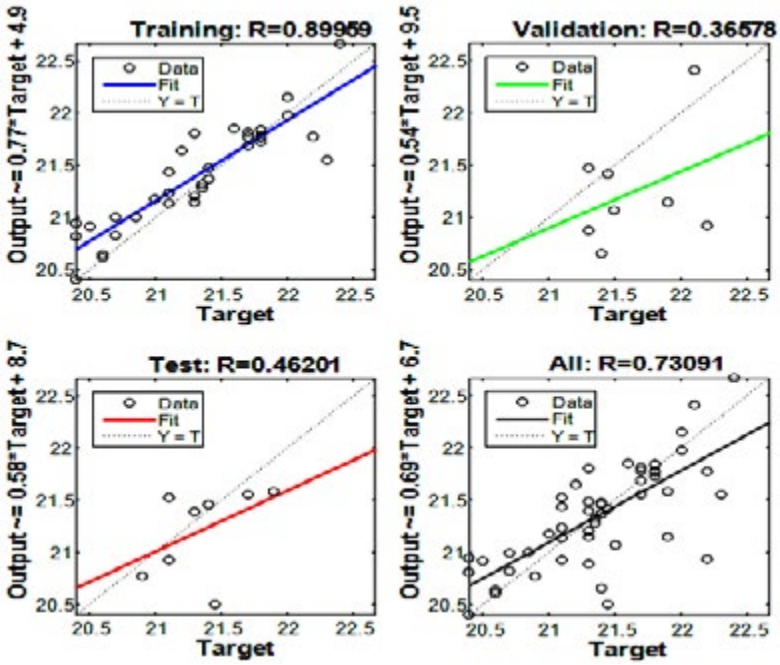

Figure 7: Regressions Coefficients

The way of perceiving information here is easier from the point of view of the type of process being perceived. For processes changing in time, it is better to use neural networks with a recurrent structure, and when there are timing chains, complex issues are considered.

When testing all the vectors with which the system was trained, the average arithmetic error was 0.89 and the average deviation was 0.73. The minimum and maximum errors of all vectors are determined and then averaged arithmetically.In our case, this results in 1.295.

The values quoted above show the accuracy of the training of the neural network and its adequacy and show the capabilities

of artificial intelligence (in particular artificial neural networks) to perceive information and adapt it. In the specific case, the ANN applied proved to be able to predict the value of EPFRs based on five other parameters (MLP: O3 (mg/m3), CO (mg/m3), NO2(µg/m3), SO2(µg/m3), PM2.5 (µg/m3)). From a practical point of view, the proposed method is valuable, as based on some commonly measured parameters, the value of EPFR can be predicted. All this shows the capabilities of artificial intelligence (in particular, artificial neural networks) to perceive information and adapt it. One of the possible steps in aiming to improve the model's accuracy of the prediction is to increase the data on the changes of the individual parameters (currently there are 51).

4. Conclusions

Either electron spin resonance (ESR) or electron paramagnetic resonance spectroscopy (EPR) is the method used most frequently to measure EPFRs. Studies on the atmospheric EPFR concentrations and degradation patterns have effectively used both methodologies. However, these methods have some drawbacks related mainly to the uncertainty of solvent extraction as a method that guarantees that all EPFRs can be efficiently extracted from the sample. Additionally, measuring EPFR by EPR or ESR is difficult since it calls for specialized equipment and skilled workers. The core idea of the paper is to rely on a database of the routinely measured air quality parameters (air pollutants O3, CO, NO2, SO2, PM2.5) and corresponding EPFR data to develop a model for EPFR prediction. One of the modern means of determining the values of free radicals is modern information technology. The so-called intelligent instruments can "adaptively" perceive the pollution information and, on this basis, obtain values for EPFR without direct measurement. Applying the above approach, an average deviation of 0.73 was obtained. One of the possible steps in aiming to improve the model's accuracy of the prediction is to expand the database of individual parameter changes towards the EPFR values.

Author Contributions: Conceptualization, V.N, S.S. and H.Y.; methodology, E.S. and V.N; software, S.S. and E.S.; validation, Y.M and F.Z..; resources, V.N. and S.S.; writing—original draft preparation, V.N. and S.S.; writing—review and editing, H.Y. and V.N.; visualization, E.S.; supervision, V.N. and S.S.; funding acquisition, S.S. and V.N. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data that support the findings of this study are available upon request from the corresponding author.

Conflicts of Interest: The authors declare that there is no conflict of interest regarding the publication of this paper.

Appendix

O_3	CO	NO_2	SO_2	PM2.5	EPFRs
35.00	0.80	40.00	7.00	40.00	21.30
50.00	0.75	48.00	3.50	100.00	22.20
65.00	0.50	35.00	2.50	20.00	22.30
35.00	0.90	60.00	7.50	75.00	21.80
60.00	1.40	65.00	9.00	90.00	20.70
50.00	1.60	80.00	10.00	100.00	20.60
75.00	1.00	65.00	6.50	75.00	21.70
52.00	1.40	97.00	11.00	170.00	20.40
50.00	0.80	78.00	5.00	65.00	21.60
60.00	0.55	48.00	6.00	48.00	22.20
75.00	1.80	50.00	7.00	220.00	21.80
73.00	0.30	34.00	2.00	22.00	21.30
50.00	0.25	35.00	4.50	18.00	21.20
45.00	1.10	63.00	15.00	27.00	21.30
76.00	0.80	49.00	8.00	25.00	21.10
60.00	0.40	40.00	3.50	11.00	21.70
90.00	0.71	34.00	3.10	94.00	21.10

95.00	0.23	43.00	1.90	13.00	22.10
110.00	0.50	32.00	7.00	81.00	21.80
130.00	1.20	48.00	5.00	145.00	21.70
112.00	0.45	38.00	2.30	35.00	22.40
134.00	0.74	51.00	7.60	53.00	21.40
59.00	0.38	23.00	1.90	48.00	21.90
135.00	0.81	47.00	5.10	19.00	21.10
142.00	0.75	31.00	2.40	63.00	21.00
74.00	0.80	23.00	2.10	21.00	21.10
51.00	0.80	53.00	6.30	12.00	20.50
60.00	0.78	45.00	2.50	11.00	20.70
32.00	0.30	22.00	0.20	8.00	21.30
46.00	0.20	18.00	0.19	13.00	21.40
38.00	0.75	67.00	4.70	148.00	21.35
34.00	1.10	64.00	2.30	46.00	20.85
43.00	0.45	49.00	2.40	112.00	20.40
41.00	1.24	78.00	5.10	82.00	21.40
37.00	0.45	68.00	6.30	12.00	21.35
43.00	0.23	18.00	2.10	18.00	21.45
27.00	0.81	32.00	2.80	181.00	21.30
18.00	0.43	41.00	2.10	78.00	21.50
23.00	0.87	52.00	4.30	141.00	21.30
22.00	0.89	63.00	4.40	28.00	20.40
20.00	1.56	65.00	4.60	25.00	21.80
45.00	0.75	43.00	0.37	49.00	21.10
19.00	0.90	53.00	0.69	43.00	21.90
12.00	1.10	47.00	8.90	203.00	20.60
45.00	1.80	54.00	7.20	61.00	20.90
21.00	1.10	63.00	12.70	191.00	21.40
18.00	2.40	84.00	9.80	87.00	21.45
11.00	1.20	63.00	11.00	25.00	22.00
35.00	0.90	60.00	7.50	75.00	21.80
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27.00	0.81	32.00	2.80	181.00	21.30
18.00	0.43	41.00	2.10	78.00	21.50
23.00	0.87	52.00	4.30	141.00	21.30
22.00	0.89	63.00	4.40	28.00	20.40
20.00	1.56	65.00	4.60	25.00	21.80
45.00	0.75	43.00	0.37	49.00	21.10
19.00	0.90	53.00	0.69	43.00	21.90
12.00	1.10	47.00	8.90	203.00	20.60
45.00	1.80	54.00	7.20	61.00	20.90

21.00	1.10	63.00	12.70	191.00	21.40
18.00	2.40	84.00	9.80	87.00	21.45
11.00	1.20	63.00	11.00	25.00	22.00
46.00	0.36	81.00	13.80	23.00	21.70
37.00	1.60	73.00	11.00	32.00	21.80
13.00	0.80	80.00	12.00	45.00	22.00
12.00	1.10	47.00	8.90	203.00	20.60
45.00	1.80	54.00	7.20	61.00	20.90
21.00	1.10	63.00	12.70	191.00	21.40
18.00	2.40	84.00	9.80	87.00	21.45
11.00	1.20	63.00	11.00	25.00	22.00
35.00	0.90	60.00	7.50	75.00	21.80
60.00	1.40	65.00	9.00	90.00	20.70
50.00	1.60	80.00	10.00	100.00	20.60
75.00	1.00	65.00	6.50	75.00	21.70
52.00	1.40	97.00	11.00	170.00	20.40
50.00	0.80	78.00	5.00	65.00	21.60
60.00	0.55	48.00	6.00	48.00	22.20
75.00	1.80	50.00	7.00	220.00	21.80
73.00	0.30	34.00	2.00	22.00	21.30
50.00	0.25	35.00	4.50	18.00	21.20
45.00	1.10	63.00	15.00	27.00	21.30
76.00	0.80	49.00	8.00	25.00	21.10
60.00	0.40	40.00	3.50	11.00	21.70
90.00	0.71	34.00	3.10	94.00	21.10
95.00	0.23	43.00	1.90	13.00	22.10
60.00	1.40	65.00	9.00	90.00	20.70
50.00	1.60	80.00	10.00	100.00	20.60
75.00	1.00	65.00	6.50	75.00	21.70
52.00	1.40	97.00	11.00	170.00	20.40
50.00	0.80	78.00	5.00	65.00	21.60
60.00	0.55	48.00	6.00	48.00	22.20
75.00	1.80	50.00	7.00	220.00	21.80
73.00	0.30	34.00	2.00	22.00	21.30
50.00	0.25	35.00	4.50	18.00	21.20
45.00	1.10	63.00	15.00	27.00	21.30
76.00	0.80	49.00	8.00	25.00	21.10
60.00	0.40	40.00	3.50	11.00	21.70
90.00	0.71	34.00	3.10	94.00	21.10
95.00	0.23	43.00	1.90	13.00	22.10
110.00	0.50	32.00	7.00	81.00	21.80
130.00	1.20	48.00	5.00	145.00	21.70
112.00	0.45	38.00	2.30	35.00	22.40

134.00	0.74	51.00	7.60	53.00	21.40
59.00	0.38	23.00	1.90	48.00	21.90
135.00	0.81	47.00	5.10	19.00	21.10
142.00	0.75	31.00	2.40	63.00	21.00
74.00	0.80	23.00	2.10	21.00	21.10
51.00	0.80	53.00	6.30	12.00	20.50
60.00	0.78	45.00	2.50	11.00	20.70
32.00	0.30	22.00	0.20	8.00	21.30
46.00	0.20	18.00	0.19	13.00	21.40
38.00	0.75	67.00	4.70	148.00	21.35
34.00	1.10	64.00	2.30	46.00	20.85
43.00	0.45	49.00	2.40	112.00	20.40
41.00	1.24	78.00	5.10	82.00	21.40
37.00	0.45	68.00	6.30	12.00	21.35
43.00	0.23	18.00	2.10	18.00	21.45
27.00	0.81	32.00	2.80	181.00	21.30
18.00	0.43	41.00	2.10	78.00	21.50

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